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IMPROVED PARAMETERIZATIONS FOR SURFACE RESISTANCE TO GASEOUS DRY DEPOSITION IN REGIONAL-SCALE, NUMERICAL MODELS

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16. ABSTRACT

Methods for estimating the dry deposition velocities of atmospheric gases in the United States and surrounding areas have been improved. The improvements have been incorporated into a revised module of computer coding for use in numerical models of atmospheric transport and deposition of pollutants over regional scales. The dry deposition module computes deposition velocities for five seasonal categories and 11 land use types specified in a land use map. The key improvement is the computation of bulk surface resistances according to three distinct pathways of mass transfer: to the upper portions of vegetative canopies; to the lower portions of canopies or structures; and to the ground (or water surface). With the surface resistances divided explicitly into several pathways, the bulk surface resistances for a large number of gases can be computed, if estimates of the effective Henry's Law constants and appropriate measures of the chemical reactivity of the various substances are known. This has been accomplished successfully for H2O2, HCHO, acetaldehyde, methyl hydroperoxide, peroxyacetic acid, HCOOH, NH3, PAN, and HNO2, in addition to the original species (SO2, O3, NOx, and HNO3). Other factors considered include surface temperature, stomatal response to environmental parameters, the wetting of the surface by dew or rain, and the covering of surfaces by snow. Surface emission of gases and variations of the uptake characteristics by individual plant species are not explicitly considered.

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ABSTRACT

Methods for computing the bulk surface resistances to uptake of gases have been developed for use in numerical models of atmospheric transport and deposition over regional scales. The surface resistances, which must be evaluated to compute dry deposition velocities, are broken down into three major groups that represent distinct pathways of mass transfer: to the upper portions of vegetative canopies, to the lower portions of canopies or structures, and to the ground (or water surface). The procedures for summing the pathway resistances to obtain the overall bulk surface resistances are designed for use in a module of computer coding on dry deposition. The module formulations are tailored for use with five seasonal categories and a landuse map of the United States and southern Canada that contains 11 landuse types. The equations for each resistance pathway are based primarily on the experimental information available on resistances to SO_2 and O_3 uptake. With knowledge of effective Henry's Law constants and chemical reactivity of various substances relative to the corresponding values for SO_2 and O_3, the surface resistances for other substance can be estimated. These substances include NO_2, NO, HNO_3, methyl hydroperoxide (to represent organic peroxides), HCOOH (to represent organic acids), NH_3, PAN, and HNO_2. The results generally conform to expectations, with uptake through the stomata of leaves controlling the surface resistance of lush vegetation if the value of the effective Henry's Law constant is near that for SO_2 or if the substance is considered at least slightly reactive. Hydrogen peroxide is the only substance of those considered that is strong in both of these removal properties. Otherwise, either solubility or reactivity tends to dominate. For extremely soluble substances (with very large effective Henry's Law constant) such as HNO_3, all exposed surfaces are computed to have very small resistances. It is necessary, however, to consider the gas-phase physical resistances because they can strongly limit transport inside canopies. Other factors considered include surface temperature, stomatal response to environmental parameters, the wetting of surfaces by dew and rain, and covering of surfaces by snow. Surface emissions of gases, the possibility of "compensation" points in biota, and variations of uptake characteristics by individual plant species within landuse types are not taken explicitly into account.

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SECTION 1

INTRODUCTION

The removal of gases from the atmosphere by turbulent transfer and uptake at the surface provides a primary means of cleansing the atmosphere and delivering chemical doses to surface components. This process of dry deposition is important in a number of environmental issues, particularly "acid rain," which deals with transport, chemical transformations, and deposition over regional or continental scales. Numerical simulations of such phenomena are often necessary to evaluate the effects of emissions in one area on deposition in another. The concern here is with dry deposition parameterizations suitable for use in numerical simulations.

The dry deposition module that has been used with the Regional Acid Deposition Model (NCAR, 1985, 1986) provides methods to compute the dry deposition velocities for SO_2, O_3, NO_x (defined here as the sum of NO and NO_2), sulfate in submicron particles, and HNO_3 in the United States and southern Canada (Sheih *et al.*, 1986; Walcek *et al.*, 1986). The basis for the parameterizations of the deposition velocities are micrometeorological formulas and, for SO_2, O_3, and NO_x, tables of resistances to uptake. The main purpose of this report is to put these tables on a more solid scientific footing, within a framework that enables logical extension to other gaseous substances. For the Regional Acid Deposition Model, these other substances include hydrogen peroxide, aldehydes, organic peroxides, peroxyacetic acid, organic acids, ammonia, and peroxyacetyl nitrate. Improvements in deposition velocities for particulate substances are not addressed here.

Another improvement needed in dry deposition calculations is a more explicit means to evaluate changes in surface resistances caused by the effects of surface wetness associated with dew and rain. Sheih *et al.* (1986) suggested that the uptake of O_3 is inhibited by dew while the uptake of SO_2 may be enhanced for approximately two hours after dewfall if both dew and good atmospheric mixing are present. For surfaces wetted by rain, Sheih

et al. implied that the surface resistance to SO_2 may increase while uptake of O_3 may be unaffected. In this report, further recommendations are given that will be incorporated into parameterizations for computing surface resistances for several of the gases of interest.

SECTION 2

FRAMEWORK OF SURFACE RESISTANCE FORMULATIONS FOR SO_2 AND O_3

The familiar framework used in past dry deposition modules relies on the simple formulation for gas deposition velocity as follows:

$$v_d \equiv -F_c/C_z, \tag{1a}$$

where F_c is the flux density and C_z is the concentration at height z. The magnitude of the deposition velocity can be found as

$$|v_d| = (r_a + r_b + r_c)^{-1}, \tag{1b}$$

where r_a is the aerodynamic resistance (common to all gases) between a specified height and the surface, r_b is the quasilaminar sublayer resistance (whose only dependence on the properties of the gas of interest is its molecular diffusivity in air), and r_c is the bulk surface resistance (e.g., Wesely and Hicks, 1977; Garland, 1977; Baldocchi *et al.*, 1987). The absolute value of v_d is taken in Equation (1b) because it can be negative and all terms on the right-hand side are constrained to being nonnegative. We assume that r_a and r_b will be evaluated as before but will suggest new methods to estimate r_c for the five seasonal categories and 11 landuse types (Table 1) employed with the module (Sheih *et al.*, 1979; Sheih *et al.*, 1986; Walcek *et al.*, 1986). The tables of r_c for SO_2 and O_3 used as primary module inputs in the past will be replaced with more detailed information.

The approach taken here is to break up the resistance r_c into several components, as is commonly done in resistance models in which series and parallel resistances are identified for various parts of the canopy (e.g., see reviews by Unsworth, 1980; Hosker and Lindberg, 1982; Baldocchi *et al.*, 1987). These components are illustrated in Fig. 1, beneath the resistances r_a and r_b. Analogously to Ohm's law in electrical circuits, r_c can be found as

$$r_c = [1/(r_s+r_m) + 1/r_{lu} + 1/(r_{dc}+r_{cl}) + 1/(r_{ac}+r_{gs})]^{-1}. \tag{2}$$

Table 1. Landuse types used to describe surface conditions in the module. In the computerized landuse map, the percentage of coverage by each of these types is given for each increment of 1/4 and 1/6 deg, longitude and latitude, respectively, from 52 to 134 deg west longitude and from 24 to 55 deg north latitude.

Type	Description
1	Urban land, almost no vegetation
2	Agricultural land, usually well watered
3	Range land, usually with low soil moisture
4	Deciduous forest
5	Coniferous forest
6	Mixed forest including wetland
7	Water, including both salt and fresh water
8	Barren land, mostly desert
9	Non-forested wetland
10	Mixed agricultural and range land
11	Rocky open areas occupied by low-growing shrubs

This formulation assumes that the concentrations representative of the plant mesophyll (C_m), plant tissue of leaves in the upper canopy (C_{lu}), substrates in the lower canopy (C_{cl}), and substrates at the ground surface (C_g) are in equilibrium with the concentration (C_z) in air (e.g., by Henry's Law constant). In this work, all these substrate concentrations will be assumed to be zero. However, it may be useful to use nonzero values for gases such as NO, NO_2, and NH_3 that are emitted from the surface, as will be discussed later.

The resistances depicted in Fig. 1 as contributing to r_c require some general explanation. First, they represent bulk properties and are not usually simply related to a single measurable quantity in the field. Rather, they correspond to properties or behaviors inferred from measurements of net vertical fluxes above the bulk surface. For example, r_s is the bulk canopy stomatal resistance and itself consists of series and parallel pathways of mass transfer due to the diffusion through leaf stomatal apertures that may exist on one or both sides of the leaves. Further, these leaves are distributed throughout the canopy and do not have uniform stomatal resistances

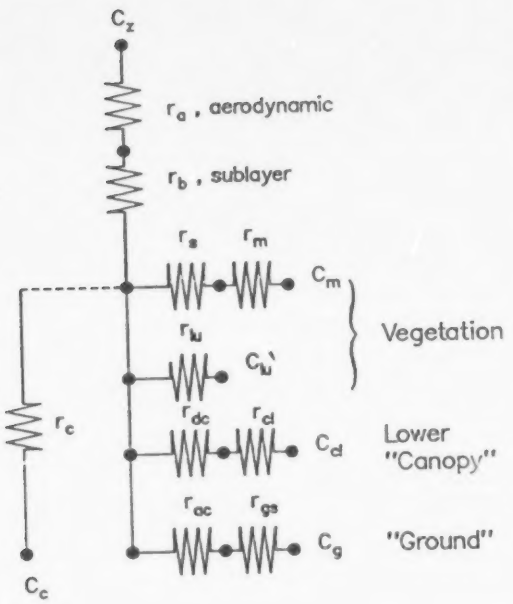

Figure 1. Schematic diagram of pathway resistances used in the module, where r_c is derived via Equation (2).

because of varying solar irradiation, temperature, ventilation, etc. The mesophyll resistance (bulk: r_m) and the resistance (bulk: r_{lu}) of the outer surfaces of leaves in the upper canopy would also vary according to position in the canopy. Another important point is that the landuse types represent very broad categories. For example, one type represents all agricultural crops, regardless of their heights and physiological characteristics. This lack of detail may result in a poor estimate of surface resistance for any particular surface cover, at a specific location and time.

The values assumed for baseline resistances are listed in Table 2. They form the basis for estimating the resistances for all the gases to be considered here. The values for r_j represent the minimum bulk canopy stomatal resistances for water vapor. It is well known that stomatal resistance varies with solar radiation and temperature. In the previous dry

Table 2. Input resistances (s m^{-1}) to the module for computations of surface resistances (r_c). Entries of 9999 indicate that there is no air-surface exchange via that resistance pathway.

Resistance component	Landuse type 1	2	3	4	5	6	7	8	9	10	11
Seasonal category 1: midsummer with lush vegetation											
r_i	9999.	60.	120.	70.	130.	100.	9999.	9999.	80.	100.	150.
r_{lu}	9999.	2000.	2000.	2000.	2000.	2000.	9999.	9999.	2500.	2000.	4000.
r_{ac}	100.	200.	100.	2000.	2000.	2000.	0.	0.	300.	150.	200.
r_{gsS}	400.	150.	350.	500.	500.	100.	0.	1000.	0.	220.	400.
r_{gs0}	300.	150.	200.	200.	200.	300.	2000.	400.	1000.	180.	200.
r_{clS}	9999.	2000.	2000.	2000.	2000.	2000.	9999.	9999.	2500.	2000.	4000.
r_{cl0}	9999.	1000.	1000.	1000.	1000.	1000.	9999.	9999.	1000.	1000.	1000.
Seasonal category 2: autumn with unharvested cropland											
r_i	9999.	9999.	9999.	9999.	250.	500.	9999.	9999.	9999.	9999.	9999.
r_{lu}	9999.	9000.	9000.	9000.	4000.	8000.	9999.	9999.	9000.	9000.	9000.
r_{ac}	100.	150.	100.	1500.	2000.	1700.	0.	0.	200.	120.	140.
r_{gsS}	400.	200.	350.	500.	500.	100.	0.	1000.	0.	300.	400.
r_{gs0}	300.	150.	200.	200.	200.	300.	2000.	400.	800.	180.	200.
r_{clS}	9999.	9000.	9000.	9000.	2000.	4000.	9999.	9999.	9000.	9000.	9000.
r_{cl0}	9999.	400.	400.	400.	1000.	600.	9999.	9999.	400.	400.	400.
Seasonal category 3: late autumn after frost, no snow											
r_i	9999.	9999.	9999.	9999.	250.	500.	9999.	9999.	9999.	9999.	9999.
r_{lu}	9999.	9999.	9000.	9000.	4000.	8000.	9999.	9999.	9000.	9000.	9000.
r_{ac}	100.	10.	100.	1000.	2000.	1500.	0.	0.	100.	50.	120.
r_{gsS}	400.	150.	350.	500.	500.	200.	0.	1000.	0.	200.	400.
r_{gs0}	300.	150.	200.	200.	200.	300.	2000.	400.	1000.	180.	200.
r_{clS}	9999.	9999.	9000.	9000.	3000.	6000.	9999.	9999.	9000.	9000.	9000.
r_{cl0}	9999.	1000.	400.	400.	1000.	600.	9999.	9999.	800.	600.	600.
Seasonal category 4: winter, snow on ground and subfreezing											
r_i	9999.	9999.	9999.	9999.	400.	800.	9999.	9999.	9999.	9999.	9999.
r_{lu}	9999.	9999.	9999.	9999.	6000.	9000.	9999.	9999.	9000.	9000.	9000.
r_{ac}	100.	10.	10.	1000.	2000.	1500.	0.	0.	50.	10.	50.
r_{gsS}	100.	100.	100.	100.	100.	100.	0.	1000.	100.	100.	50.
r_{gs0}	600.	3500.	3500.	3500.	3500.	3500.	2000.	400.	3500.	3500.	3500.
r_{clS}	9999.	9999.	9999.	9000.	200.	400.	9999.	9999.	9000.	9999.	9000.
r_{cl0}	9999.	1000.	1000.	400.	1500.	600.	9999.	9999.	800.	1000.	800.
Seasonal category 5: transitional spring with partially green short annuals											
r_i	9999.	120.	240.	140.	250.	190.	9999.	9999.	160.	200.	300.
r_{lu}	9999.	4000.	4000.	4000.	2000.	3000.	9999.	9999.	4000.	4000.	8000.
r_{ac}	100.	50.	80.	1200.	2000.	1500.	0.	0.	200.	60.	120.
r_{gsS}	500.	150.	350.	500.	500.	200.	0.	1000.	0.	250.	400.
r_{gs0}	300.	150.	200.	200.	200.	300.	2000.	400.	1000.	180.	200.
r_{clS}	9999.	4000.	4000.	4000.	2000.	3000.	9999.	9999.	4000.	4000.	8000.
r_{cl0}	9999.	1000.	500.	500.	1500.	700.	9999.	9999.	600.	800.	800.

deposition module, solar radiation categories were used to form tables of resistance, and temperature was not explicitly included (Sheih *et al.*, 1986; Walcek *et al.*, 1986). Here we use the following generalized function to estimate the bulk canopy stomatal resistance:

$$r_s = r_i\{1 + [200(G+0.1)^{-1}]^2\}\{400[T_s(40-T_s)]^{-1}\}, \quad (3)$$

where G is the solar irradiation in watts per square meter and T_s is the surface air temperature in degrees Celsius between 0 and 40 °C. Outside this range, we set r_s to a very large value, to implement the assumption that the transfer through stomata is stopped. Equation (3) is derived from information provided by Baldocchi *et al.* (1987), who reviewed a number of scientific studies to formulate similar relationships specific to certain plant species. While our landuse map does not provide the detailed information necessary to make species-specific estimates of stomatal resistance, Equation (3) should provide representative variations caused by solar radiation and temperature.

The combined minimum stomatal and mesophyll resistance for substance x can be found as

$$r_{smx} = r_s D_{H_2O}/D_x + r_{mx}, \quad (4)$$

where D_x is the molecular diffusivity of gas x in air, D_{H_2O} is the molecular diffusivity for water vapor, and r_{mx} is the mesophyll resistance for the gas of interest. For O_3 and SO_2, the mesophyll resistance is practically zero, and suggestions will be made later for methods to estimate r_{mx} for other substances.

The lower canopy resistance r_{cl} is meant to account for uptake pathways at the leaves, twig, bark, etc., while the ground surface resistance r_{gs} signifies uptake at the "ground" by soil, leaf litter, snow, water, etc. In Table 2, separate values for SO_2 and O_3 are indicated by the additional subscripts "S" and "O," respectively. We expect that all surfaces represented by r_{lc} and r_{gs} have a reduced capacity for uptake when cold. For example, surface resistances for uptake of HNO_3, SO_2, and NO_2 by snow increase markedly when its temperature decreases below 2 °C (Johansson and Granat, 1986; Valdez,

1987). Accordingly, we approximate such an effect for all surfaces by adding the value, in seconds per meter, of $1000\exp[-T_s-4]$ to r_{lc} and r_{gs} (and r_{lu}) for all substances. It is unknown whether this procedure is correct for every substance, and there may be notable exceptions. For example, recent work has shown that the rate of uptake of H_2O_2 by ice is not reduced when the temperature drops below freezing (Lee Y.N., Brookhaven National Laboratory, private communication).

The values of r_{ac} are estimated solely by landuse and season, mostly on the basis of the depth of the structure, which is usually a plant canopy. The value of r_{ac} increases with representative heights and densities of the "canopies," in accordance with findings concerning ozone deposition velocity (Wesely, 1983). The resistance r_{dc} overlaps somewhat with r_{ac} and is meant to incorporate the effects of mixing forced by buoyant convection when sunlight heats the ground or lower canopy and by penetration of wind into canopies on the sides of hills. The following simplistic formula is used:

$$r_{dc} = 100[1 + 1000(G+10)^{-1}](1 + 1000\theta)^{-1}, \qquad (5)$$

where θ is the slope in radians of the local terrain, and r_{dc} is expressed in units of seconds per meter. The value of 1000 for the coefficient of G is a rough estimate based on observations (Wesely, 1983; Wesely *et al.*, 1983); the coefficient of 1000 for θ is a first approximation for which no relevant data are at hand.

To be more realistic, part of r_{dc} should be incorporated into r_{ac} so that the latter could also be influenced by the effects of the enhanced mixing. For simplicity, however, we have chosen to incorporate all the effects into one term, anticipating that the resistance sum ($r_{dc}+r_{ac}$) may at times have to be adjusted to represent a pathway or lack of a pathway to uptake at the ground. Also, since r_{dc} is often small regardless of landuse type and seasonal category, the values of r_{cl} must be carefully chosen. For example, while r_{ac} is set to zero for lakes and bare ground, r_{dc} is unaffected but its pathway (Fig. 1) for mass transport is cut off because r_{cl} is set to a very large value for these surfaces (Table 2). Another example occurs for the seasonal category 4 (wintertime with snow) and landuse type 2 (agricultural

land) for ozone, in which r_{clO} is allowed to be as small as 1000 s m^{-1} in order to allow ozone destruction at the surfaces of dry materials such as bushes that protrude above the snow.

The gas-phase sublayer resistance r_b is shown as being common to all lower branches of the pathways in Fig. 1, although it is normally only applicable to transfer at the upper portions of the overall surface. Formulation of bulk sublayer resistances in the lower canopy and at the ground beneath the canopy is quite difficult, because these resistances should depend on local rates of momentum transfer, for which suitable parameterizations are not readily available. As a result, the approach represented by Fig. 1 may be inadequate for situations in which the molecular diffusivity of the substance of interest is much different from that for SO_2 and O_3, when exchange at surfaces below the upper canopy comprises a large portion of the net vertical flux above the canopy.

SECTION 3

EXTENSION TO OTHER SUBSTANCES

Nitric oxide, nitrogen dioxide, and nitric acid vapor were treated fairly explicitly by Sheih *et al.* (1986), but other compounds such as NH_3, H_2O_2, HCHO, other aldehydes, organic acids, organic peroxides, and peroxyacetic acid have been treated in a less thorough fashion (Chang *et al.*, 1987). As a result of their reactivity or solubility, all these substances and others such as PAN and HNO_2 may be deposited on the surface of the Earth at a significant rate. Table 3 lists the properties we will use to estimate the surface resistances for these gases. As shown in Equation (4), the ratio of their molecular diffusivities in air to that of water vapor directly affects the stomatal resistance.

The effective Henry's Law constants in Table 3 are used to estimate uptake by moist and wet surfaces, including extracellular water in the substomatal cavities of plant leaves. To represent the ability of the gas to go into such aqueous solutions, this effective Henry's Law constant incorporates the effects of acid-base equilibria at a pH of seven. While this pH may be too high for some fresh water lakes, it is reasonable for ocean water and some large lakes such the Great Lakes. Also, near-neutral conditions are typical of plant sap, whose pH presumably is not much different from that encountered in the substomatal cavities in the leaves of many plants (Canny, 1984). We assume that the hydration and formation of ions are rapid (to the extent that they occur), as is the case for SO_2 (Martin, 1984).

The values listed in Table 3 for the negative log of electron activity for half redox reactions in neutral aqueous solutions [$pe^o(W)$] and the overall second-order reaction rates with S(IV) [$k^{(2)}$] provide a means to estimate a reactivity factor f_0 for oxidation of biological substances. Three categories are used: highly reactive (f_0=1), which implies that the gas is as reactive as O_3; slightly reactive (f_0=0.1); and nonreactive (f_0=0). The slightly

Table 3. Gases considered in the dry deposition module and their properties relevant to estimating resistances to dry deposition.

Gaseous species	Symbol	D_{H_2O}/D_x[a]	H^*[b,c] ($M\ atm^{-1}$)	$pe^o(W)$[b,d]	$k(2)$[e] ($M^{-1}\ s^{-1}$)	f_0
Sulfur dioxide	SO_2	1.9	$1X10^5$	-5	-	0
Ozone	O_3	1.6	0.01	28	$6X10^8$	1
Nitrogen dioxide	NO_2	1.6	0.01	--	$2X10^6$	0.1
Nitric oxide	NO	1.3	$2X10^{-3}$	--	$<1X10^{-2}$	0
Nitric acid vapor	HNO_3	1.9	$1X10^{14}$	7	<1X10-2	0
Hydrogen peroxide	H_2O_2	1.4	$1X10^5$	23	7	1
Acetaldehyde	(ALD)[f]	1.6	15	neg.	-	0
Formaldehyde	HCHO	1.3	$6X10^3$	-3	-	0
Methyl hydroperoxide	(OP)[f]	1.6	240	--	2	0.1
Peroxyacetic acid	PAA	2.0	540	--	600	0.1
Formic acid	(ORA)[f]	1.6	$4X10^6$	-8	-	0
Ammonia	NH_3	1.0	$2X10^4$	-	-	0
Peroxyacetyl nitrate	PAN	2.6	3.6	--	$>3X10^3$	0.1
Nitrous acid	HNO_2	1.6	$1X10^5$	6	$4X10^{-4}$	0.1

-: Not relevant.

--: Information not found.

[a]Computed as the square root of ratio m_x/m_{H_2O} of molecular weights.

[b]Effective values for water with near-neutral pH.

[c]Drawn primarily from summaries by Schwartz (1984) and Gaffney *et al.* (1987); Lind and Kok, 1986 (methyl hydroperoxide and PAA); Jacob, 1986 (formic acid).

[d]Obtained primarily from Stumm and Morgan (1981) and Morel (1983).

[e]Pseudo-first-order rate constants for oxidation of S(IV) in water with near-neutral pH, derived for O_3 (Hoffman, 1986); NO_2 (Lee and Schwartz, 1983); NO and HNO_3 (Martin *et al.*, 1981); H_2O_2 (articles reviewed by Seinfeld, 1986); methyl hydroperoxide and PAA (Lind *et al.*, 1987); PAN (Lee, 1984; Calvert *et al.*, 1985); HNO_2 (Oblath *et al.*, 1981).

[f]Class of substances (aldehydes, organic peroxides, and organic acids) represented by the species shown.

reactive category is intended primarily to set plant leaf mesophyll resistances to very small values and to allow rapid uptake of gases through plant leaf stomata. As is evident in Table 3, the value of f_0=0.1 is assigned to substances that have a rather small value of $pe^o(W)$ but still have a significant second-order reaction with S(IV). This procedure is based on the hypothesis that if a substance can oxidize S(IV), it will be removed rapidly in the slightly reducing environment of the substomatal materials. Support for this hypothesis comes from the observations that NO_2 seems to have a very small mesophyll resistance (Wesely *et al.*, 1982; Delaney and Davis, 1983). The use of reactivity with S(IV) as an indicator, nevertheless, is only an expedient that takes advantage of the large amount of information that has become available on this pseudo-first-order reaction rate (see references listed in Table 3). This is not meant to imply that the S(IV) components of $SO_2(aq)$, HSO_3^-, and SO_3^{2-} in extracellular water are unaffected by other reactions [e.g., see Mudd (1975a) for a discussion of the fate of SO_2]. The reality may be that the pathways of uptake of substances such as NO_2 are strongly influenced by enzymatic processes (e.g., Law and Mansfield, 1982). Further, these pathways may be cut off when the gas-phase concentrations reach a fairly low value corresponding to a compensation point for the biota (Johansson, 1987).

The mesophyll resistance in Equation (4) is computed for any substance x on the basis of the key parameters H^* and f_0:

$$r_{mx} = (H^*/3000 + 100f_0)^{-1}. \qquad (6)$$

This relationship assumes that two parallel pathways exist to the extracellular water inside plant leaf stomata. The pathway based on f_0 produces very small values of r_{mx} when f_0 is 1 or 0.1. The other pathway is via dissolution in the aqueous solution, and the factor 3000 M atm^{-1} is derived in part from the consideration that if H^* is sufficiently large, the concentration that can be achieved in the aqueous phase represents a large sink for the substance. As Equation (6) indicates, values of H^* greater than about ten will result in fairly rapid uptake. The exact value of 3000 is derived from considerations of CO_2 uptake by vegetation. For example, it is well known that uptake rates of 1 mg m^{-2} s^{-1} are approached with lush

vegetation in optimum ambient conditions (e.g., Monteith, 1973). This rate corresponds to a (bulk canopy) mesophyll resistance of approximately 600 s m^{-1}. At a pH of seven, H* for CO_2 is near 4.4, and the value of 3000 as a rounded-off equivalent to (4.4)(600) is used in Equation (6). Although the hydration rate of CO_2 is normally slow (Quinn and Otto, 1971), we have assumed that it is accelerated by enzymatic reactions in the leaf, which would allow rapid equilibrium to be established.

The resistance of the outer surfaces of leaves in the upper canopy is computed as

$$r_{lux} = r_{lu}(10^{-5}H^* + f_0)^{-1}. \qquad (7)$$

This allows for two parallel routes to the substrate, one analogous to SO_2 pathways and one to O_3 pathways. Substances such as HNO_3 that have a very high solubility in aqueous solution will be removed rapidly at the surface, as is indicated by the first term on the right-hand side. The second term allows highly reactive substances to behave as O_3 and is effective in reducing uptake for substances classified as slightly reactive (f_0=0.1).

For uptake of a gas that has a nonzero value of f_0 and does not have an extremely large value of H*, Equation (7) implies that the gas will diffuse though the outer surfaces of leaves in the same way as do SO_2 and O_3. No adjustments are made for molecular size and varying diffusivities in air, just as no differentiation between SO_2 and O_3 is made in the values of r_{lu}. We could, for example, hypothesize that uptake at outer surfaces is limited by molecular diffusion in air and then modify the terms on the right-hand side accordingly. This hypothesis would envision the outer surfaces as being mostly impervious except for tiny air-filled gaps, smaller than stomatal openings. However, such scaling with diffusivities is questionable even for CO_2, which is one of the few substances other than water vapor that has been studied with regard to leaf cuticular resistances (e.g., Holmgren *et al.*, 1965). In addition, Equation (7) does not account for the possibility that nonpolar substances might dissolve readily in the waxy covering of the cuticle. For example, it has been found that vapor-phase PCBs accumulate to a

significant extent in plant leaves (Buckley, 1982), and that the octanol-water partition coefficient appears to be a key parameter for describing the foliar uptake of organic compounds (Travis and Hattemer-Frey, 1988).

The resistance of the exposed surfaces in the lower portions of structures (canopies, buildings) above the ground is computed as the parallel sum of resistances corresponding to those for SO_2 and O_3:

$$r_{clx} = [H^*/(10^5 r_{clS}) + f_0/r_{cl0}]^{-1}. \quad (8)$$

The two terms on the right-hand side have the same function as the terms on the right-hand side of Equation (7), but here the individual resistances, r_{clS} and r_{cl0}, have been incorporated.

Computation of resistance to uptake at the "ground" surface takes an approach equally simple as that in Equations (7) and (8). For the varied situations that include bodies of water, bare soil, and litter beneath forest canopies, the resistance is computed as

$$r_{gsx} = [H^*/(10^5 r_{gsS}) + f_0/r_{gs0}]^{-1}. \quad (9)$$

For surfaces with a pH significantly less than seven, it would be desirable to use appropriately adjusted values of H^*, even for SO_2.

SECTION 4

EFFECTS OF DEW AND RAIN

One effect of dew and rain at the surface is the covering of leaf stomata, which cuts off direct gas exchange when the stomata are open. Field observations in recent unpublished experiments at Argonne National Laboratory indicate that about two-thirds of the leaves in a canopy are typically covered, although the amounts can vary widely. Accordingly, the value of r_s in Equation (4) should be increased by a factor of three. The only other adjustment we make is in the value of r_{lu}, for the leaves in the upper canopy. Since we thus assume that altered resistances in other portions of the canopy are relatively insignificant, care must be taken to make arithmetic adjustments that allow some effects of dew when the "canopy" is not vegetative, such as in urban areas. For dew, the results of the experiments at Argonne National Laboratory indicate that the resistance to SO_2 uptake by vegetation is probably decreased, but the uptake of O_3 is retarded. A reasonable approximation for the resistance to SO_2 uptake by a dew-covered surface is

$$r_{luS} = 100 \text{ s m}^{-1}, \tag{10}$$

which results in a very small value of r_c. For O_3, the dew acts as a barrier to surface removal:

$$r_{luO} = [1/3000 + 1/(3r_{lu})]^{-1}, \tag{11}$$

where the second term on the right-hand side takes into account the reduction of dry areas available for removal. The value of 3000 s m^{-1} in the first term is only a rough approximation.

Wetting of surfaces by rainfall probably has quite different effects. Unpublished measurements by Argonne National Laboratory indicate that rainfall results in an increase of resistance to SO_2 uptake and may actually decrease the resistance to O_3. This is reasonable because the rainfall may be sufficiently saturated with S(IV) to prevent further uptake of SO_2 and

allow limited chemical reaction with O_3. The extent of such behavior would depend on the chemical properties of the ambient atmosphere and of the substrates, as well as those of the rainfall. As a first approximation, we estimate the resistance to SO_2 uptake by a surface wetted by rainfall to be

$$r_{luS} = [1/5000 + 1/(3r_{lu})]^{-1}, \tag{12}$$

which has a structure similar to Equation (11). For O_3, rainfall might have a weaker effect on surface removal:

$$r_{luO} = [1/1000 + 1/(3r_{lu})]^{-1}. \tag{13}$$

A special exception should be made for SO_2 uptake in urban areas. Preliminary results indicate that fairly pervious artificial materials such as limestone take up SO_2 rather rapidly when wet or when the relative humidity is high (Youngdahl A.C., Argonne National Laboratory, private communication). In a attempt to take this factor into account, we recommend that the value of r_{luS} be adjusted for landuse type 1 when wet conditions prevail, so that Equations (10) and (12) are then replaced with the following:

$$r_{luS} = 50 \text{ s m}^{-1}.$$

This will probably require modification as better information is obtained.

For substances other than SO_2 and O_3, the following formulation replaces Equation (7) if either dew or rain wets the surface:

$$r_{lux} = [1/(3r_{lu}) + 10^{-7}H^* + f_0/r_{luO}]^{-1}, \tag{14}$$

where reduction of dry area, solubility in water, and chemical reactivity are taken into account by the first, second, and third terms, respectively, on the right-hand side. In this case, r_{luO} is taken from Equation (11) for dew and from Equation (13) for rain.

In practical application of Equations (10) through (14), information on dewfall and rainfall events must be provided from sources external to the dry deposition module. A direct computation of the surface wetness would be most desirable, e.g., by estimating the amount of free surface water accumulated and then evaporated. Alternatively, surface relative humidity might be a useful index. After dewfall and rainfall events are completed, surface wetness often disappears as a result of evaporation after approximately two hours of good atmospheric mixing, the period of time recommended earlier (Sheih *et al.*, 1986) and found to be typical for dew during recent experiments at Argonne Natinal Laboratory. An important subcategory of surface wetness is that caused by fog or cloud water impaction on surface elements. Resistances would probably then be altered in the same fashion as they are for rainfall, since the chemistries may be similar.

SECTION 5

EXAMPLES AND DISCUSSION

The tables in Appendix I give examples of surface resistances (r_c) for selected values of solar irradiation and one case each for dew and rain when G is zero. The surface temperatures assumed were 25, 10, 2, 0, and 10 °C for seasonal categories 1-5, respectively. The values shown have been truncated to give no more than two significant digits. For SO_2 and O_3, there are some significant differences from the previous version of the dry deposition module (Sheih *et al.*, 1986), but the present results are more rigorously derived and probably more realistic. Overall, we anticipate that the deposition velocities for the two substances as derived with these procedures will be hardly distinguishable from estimates made from the previous version of the module. Likewise, little should have changed with regard to deposition velocities for NO_2, NO, and HNO_3. The latter two are not represented in Appendix I because the computed values of r_c are always very large (entry of 9999) for NO and zero for HNO_3. Neither set of surfaces resistances is very controversial, except that NO (and some of the other common nitrogen compounds) may be emitted from the surface rather than removed from the atmosphere (Galbally 1985; Anderson and Levine, 1987; Williams *et al.*, 1987) and that notable changes of HNO_3 fluxes with height can occur if HNO_3 is present (Huebert B.J., University of Rhode Island, private communication). In practical applications involving regional-scale atmospheric models, a small value of r_c near 10 s m^{-1} should be used rather than the zeros indicated in Appendix I for HNO_3 and for some other substances under certain conditions, in order to avoid unrealistic situations of extremely high deposition velocities over unusually rough surfaces.

As is desired, the calculated surface resistances for NO_2 are usually quite large except for lush vegetation exposed to solar radiation, when leaf stomata are open. This conforms to a number of experimental investigations involving atmospheric concentrations of at least a few ppbV of NO_2 over a variety of surfaces recognized in each study as not being sources of the gas

(e.g., Wesely *et al.*, 1982; Delaney and Davis, 1983; Gravenhorst and Bottger, 1983). Use of the value f_0=0.1 in Equation (6) implies fairly efficient uptake through stomata. It has been suggested that the measurement methods used in some of the field work tended to cause underestimates of NO_2 surface resistances because of interference by HNO_3 in sensor response (Huebert, 1983), but subsequent unpublished reevaluations by several investigators have indicated that overestimates were more likely because the interference may have increased only the mean concentrations and not the eddy fluctuations measured. The recommendations of Sheih *et al.* (1986), which ignored the possible HNO_3 effect, are still plausible: the minimum computed values of r_c for NO_2 should be about 1.75 times the corresponding O_3 resistances. According to the entries for seasonal category 1 for a value of G=800 W m^{-2} s^{-1} in Appendix I, the ratios of r_c for NO_2 to r_c for O_3 average to about 1.5 for the landuse types that indicate a surface covered with vegetation.

The resistances for NO_2 for surfaces other than sunlit vegetation appear to be quite large (Appendix I). This results from use of relatively small values of f_0 and H* in Equations (7), (8), and (9). Sheih *et al.* (1986) suggested that these resistances should be high, near 1000 s m^{-1}, and recommended that, in general, the sum of NO and NO_2 should be considered rather than NO_2 alone. This was suggested because rapid in-air chemical reactions can cause a significant change of NO and NO_2 vertical fluxes between the surface and the point at which deposition velocities are applied, but the sum of NO and NO_2 fluxes should be practically unchanged (Fitzjarrald and Lenschow, 1983). This summing is clearly a desirable procedure, especially in nonurban environments away from local anthropogenic emissions where concentrations of NO_2 and NO are much smaller than O_3 concentrations.

The resistances for the other substances in Appendix I are predictions that in many cases have no supporting field or laboratory observations. Hydrogen peroxide has the unusual properties of being both moderately soluble in water, as is SO_2 near a pH near seven, and as strong an oxidizing agent as O_3. Rapid removal takes place at wet surfaces and moderately rapid deposition occurs over vegetation. Many surfaces that may seem somewhat inert, such as in unharvested agricultural areas, remove H_2O_2 fairly

efficiently. Solubility alone is highlighted in the surface resistances calculated for formaldehyde (HCHO), formic acid (HCOOH, or ORA to represent organic acids), and acetaldehyde (CH_3CHO, or ALD to represent aldehydes other than HCHO). Formaldehyde is taken up rather rapidly at liquid water surfaces and by sunlit vegetation, but has much less interaction with soils and senescent vegetation. The rather large solubility of formic acid allows it to be taken up rapidly at many different types of surfaces. Variations on the same theme are seen for NH_3 and HNO_2. Laboratory and field observations have shown that NH_3 can be taken up quickly through leaf stomata and by moist surfaces in an agricultural crop (Hutchinson *et al.*, 1972; Denmead *et al.*, 1978; Lemon and Van Houtte, 1980; van Hove *et al.*, 1987). The resistances to NH_3 uptake by green vegetation (seasonal categories 1 and 5) are smaller than the corresponding resistances for SO_2 because r_{smx} calculated with Equation (4) for NH_3 is reduced by almost a factor of two as a result of its smaller molecular diffusivity.

This effort to evaluate surface resistances does not consider the fact that NH_3 is often emitted from soils, particularly when atmospheric concentrations are lower than a few ppbV (e.g., Dabney and Bouldin, 1985). As was already discussed, NO and NO_2 might also be emitted from the surface. A first approximation to account for these emissions and thus obtain more realistic estimates of air-surface exchange would involve adjusting r_c values found with Equation (2) for NH_3, NO, and NO_2 by multiplying each r_c by $C_z/(C_z-C_c)$, where C_c is equal to an assumed compensation point of 2 to 3 ppbV.

The remaining three substances, methyl hydroperoxide (CH_3O_2H, or OP to represent several organic peroxides), peroxyacetic acid [$CH_3C(O)O_2H$, or PAA], and peroxyacetyl nitrate [$CH_3(O)O_2NO_2$, or PAN], have slightly limited solubility and are moderately reactive as oxidants. PAN is the least soluble and thus has the largest surface resistance of the three substances for sunlit green vegetation. Interpretation of data of Mudd (1975b) leads to the conclusion that PAN (as well as peroxypropionyl nitrate) can be taken up quite rapidly through leaf stomata; Hill (1971) and Garland and Penkett (1976) found that deposition velocities for PAN are typically 1/2 to 1/3 of those for O_3. This is at least partially consistent with the entries in Appendix I, from which we infer that maximum deposition velocities near 0.005 m s^{-1} are

possible, for the optimum conditions of a fully sunlit, lush vegetative canopy. By comparison with the computed surface resistances for acetaldehyde, it is apparent that the chemical reactivity of PAN is probably the major factor in affecting its surface resistance, not its solubility in water.

SECTION 6

CONCLUSIONS

A scheme has been presented that breaks the surface resistances (r_c) for atmospheric gases into components that represent mass transfer pathways at the stomatal apertures of leaves, mesophyll of leaves, outer surfaces of leaves in the upper canopy, exposed surfaces in the lower portions of the canopy or structure, and "ground" surfaces that include soil, leaf litter, and water. Gas-phase resistance pathways leading to the latter two locations inside or beneath canopies are also specified. Because of the relatively large amount of information available on the dry deposition velocities of SO_2 and O_3, their resistances are estimated explicitly for the above components. Based on the solubilities and chemical reactivities relative to those for SO_2 and O_3, formulas are suggested for the surface resistance components for several other substances important in acid deposition or related atmospheric chemistry. These include NO_2, NO, HNO_3, H_2O_2, HCHO, acetaldehyde (to represent aldehydes other than HCHO), methyl hydroperoxide (to represent organic peroxides), peroxyacetic acid, HCOOH (to represent organic acids), NH_3, PAN, and HNO_2. For many of these substances, the resistances computed represent predictions that have yet to be verified with observations. This scheme can be used to predict the surface removal of many other substances as well.

The formulations used to compute the surface resistance components and add them together to obtain values of r_c for various substances and types of surfaces are derived in a form suitable for use in numerical models of regional-scale atmospheric transport, chemical transformation, and deposition. This allows updating of the dry deposition module of the Regional Acid Deposition Model (RADM). Appendix II gives a listing of the Argonne version of the dry deposition module, which may be adapted for use with RADM or other models that require the information. It should be noted, however, that the surface resistances that can be computed are specific to 11 landuse types (Table 1) and thus might not be directly useful in numerical models that use different surface characterizations.

The scheme for computing surface resistances is rather simplistic because it categorizes all surfaces by only 11 landuse types and considers five general seasonal categories. The dry deposition module will probably not produce very accurate estimates of dry deposition for a short time period at a particular small area. Rather, the estimates are intended for long-term averages over at least several weeks and for rather large areas over which the individual variations of plant species composition and factors such as soil moisture content are smoothed. For vegetation, uptake resistances by individual plant species have not been identified, and the influence of varying leaf areas (green or senescent) has not been tied explicitly to a measurable quantity such as LAI (leaf area index). The following factors are considered in a general fashion: vegetation height, aridity or soil moisture content, surface temperature, and variations of leaf stomatal resistance with solar radiation and temperature.

Wet surfaces have a variety of surface resistances, which the module generalizes considerably. For open bodies of water, we have assumed an aqueous pH near seven, which is undoubtedly too large for many small bodies of water. For ozone, we have assumed that the surface resistance suitable for seawater is relevant (r_c=2000 s m^{-1}), which is probably too small for fresh water. Uptake by vegetation wetted by dewfall is assumed to be enhanced for SO_2 and retarded for O_3, while the opposite changes are suggested for surfaces wetted by rainfall. It is likely that surface wetness caused by impaction of fog or cloud water would alter surface resistances in a manner similar to rainfall. The effects of dew, rain, fog, and cloud water, however, are not well understood, and revisions to the approach taken here might be necessary as further information becomes available.

A number of factors that can strongly influence air-surface exchange rates are not considered here. Gases that undergo rapid chemical reactions in the air should be treated cautiously because resulting changes of fluxes with height might cause unusual variations of deposition velocity with height, e.g. for O_3, NO, or NO_2 treated separately. Soil alkalinity or acidity can have a strong influence on the surface resistances of some substances; SO_2 removal rates by dry or acidic soils are undoubtedly much slower than by wet or alkaline soils. For many substances, natural emissions may be much more

important than dry deposition in affecting local air chemistry, e.g., for NO and NH_3. In addition, the use of a deposition velocity for NO_2 and NH_3 can be misleading if concentrations near the surface are not considered, because many surfaces have a compensation point corresponding to an atmospheric concentration of 2-3 ppbV, below which the surface resistance becomes very large. This could occur for other substances as well.

REFERENCES

Anderson I.C. and Levine J.S. (1987) Simultaneous field measurements of biogenic emissions of nitric oxide and nitrous oxide. *J. Geophys. Res.* **92**, 965-976.

Baldocchi D.D., Hicks B.B. and Camara P. (1987) A canopy stomatal resistance model for gaseous deposition to vegetated surfaces. *Atmospheric Environment* **21**, 91-101.

Buckley E.H. (1982) Accumulation of airborne polychlorinated biphenyls in foliage. *Science* **216**, 520-522.

Calvert J.G., Lazrus A., Kok G.L., Heikes B.G., Walega J.G., Lind J. and Cantrell C.A. (1985) Chemical mechanisms of acid generation in the troposphere. *Nature* **317** 27-35.

Canny M.C (1984) Translocation of nutrients and hormones. In *Advanced Plant Physiology* (edited by Wilkens M.B.), pp. 227-296, Pitman Publishing Inc., Marsfield, MA.

Chang J.C., Brost R.A., Isaksen I.S.A., Madronich P., Middleton P., Stockwell W.R. and Walcek C.J. (1987) A three-dimensional Eulerian acid deposition model: Physical concepts and formulation. *J. Geophys. Res.* **92**, 14,681-14,700.

Dabney S.M. and Bouldin D.R. (1985) Fluxes of ammonia over an alfalfa field. *Agron. J.* **77**, 572-578.

Denmead O.T., Nulsen R. and Thurtell G.W. (1978) Ammonia exchange over a corn crop. *Soil Sci. Soc. Am.* **42**, 840-842.

Delaney A.C. and Davis T.D. (1983) Dry deposition of NO_x to grass in rural East Anglia. *Atmospheric Environment* **17**, 1391-1394.

Fitzjarrald D.R. and Lenschow D.H. (1983) Mean concentration and flux profiles for chemically reactive species in the atmospheric surface layer. *Atmospheric Environment* **17**, 1391-1394.

Gaffney J.S., Streit G.E., Spall W.D. and Hall J.H. (1987) Beyond acid rain. *Env. Sci. Tech.* **21**, 519-524.

Galbally I.E. (1985) The emission of nitrogen to the remote atmosphere: Background paper. In *The Biogeochemical Cycling of Sulfur and Nitrogen in the Remote Atmosphere* (edited by Galloway J.N. *et al.*), pp. 27-53, D. Reidel Publishing Company, Dordrecht, Holland.

Garland J.A. (1977) The dry deposition of sulphur dioxide to land and water surfaces. *Proc. R. Soc. Lond.* **12**, 245-268.

Garland J.A. and Penkett S.A. (1976) Absorption of peroxy acetyl nitrate and ozone by natural surfaces. *Atmospheric Environment* **10**, 1127-1131.

Gravenhorst G. and Bottger A. (1983) Field measurements of NO and NO_2 fluxes to and from the ground. In *Acid Deposition* (edited by Beilke A. and Elshout A.J.), pp. 172-184, D. Reidel Publishing Company, Dordrecht, Holland.

Hill A.C. (1971) Vegetation: A sink for atmospheric pollutants. *Air Pollut. Control Ass.* 21, 341-346.

Hoffman M.R. (1986) On the kinetics and mechanism of oxidation of aquated sulfur dioxide by ozone. *Atmospheric Environment* 20, 1145-1154.

Holmgren P., Jarvis P.G. and Jarvis M.S. (1965) Resistances to carbon dioxide and water vapour transfer in leaves of different plant species. *Physiologica Plantarum* 18, 557-573.

Hosker R.P.Jr. and Lindberg S.E. (1982) Review: Atmospheric deposition and plant assimilation of gases and particles. *Atmospheric Environment* 16, 889-910.

Huebert B.J. (1983) Discussions, on "An Eddy Correlation Measurement of NO_2 Flux to Vegetation and Comparison to O_3 Flux." *Atmospheric Environment* 17, 1600.

Hutchinson G.L., Millington R.J. and Peters D.B. (1972) Atmospheric ammonia: Absorption by plant leaves. *Science* 175, 771-772.

Jacob D.J. (1986) Chemistry of OH in remote clouds and its role in the production of formic acid and peroxymonosulfate. *J. Geophys. Res.* 91, 9807-9826.

Johansson C. (1987) Pine forest: A negligible sink for atmospheric NO_x in rural Sweden. *Tellus* 39B, 426-438.

Johansson C. and Granat L. (1986) An experimental study of the dry deposition of gaseous nitric acid to snow. *Atmospheric Environment* 20, 1165-1170.

Law R.M. and Mansfield T.A. (1982) Oxides of nitrogen in the greenhouse atmosphere. In *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (edited by Unsworth M.H. and Ormrod D.P.), pp. 93-112, Butterworth Scientific, London.

Lee Y.-N. (1984) Kinetics of some aqueous-phase reactions of peroxyacetyl nitrate. In *Gas-Liquid Chemistry of Natural Waters*, Vol. 1 (edited by Newman L.), pp. 21-1 to 21-9, BNL 51757, Brookhaven National Laboratory, Upton, Long Island, New York. Available from U.S. Department of Energy Technical Information Center, Box 62, Oak Ridge, TN 37830.

Lee Y.-N. and Schwartz S.E. (1983) Kinetics of oxidation of aqueous sulfur(IV) by nitrogen dioxide. In *Precipitation Scavenging, Dry Deposition and Resuspension*, Vol. 1 (edited by Pruppacher H.R., Semonin R.G. and Slinn W.G.N.), pp. 453-466, Elsevier Science Publishing Co., Inc., New York.

Lemon E. and Van Houtte R. (1980) Ammonia exchange at the land surface. *Agron. J.* 72, 876-883.

Lind J.A. and Kok G.L. (1986) Henry's Law determinations for aqueous solutions of hydrogen peroxide, methylhydroperoxide, and peroxyacetic acid. *J. Geophys. Res.* **91**, 7889-7895.

Lind J.A., Lazrus A.L. and Kok G.L. (1987) Aqueous phase oxidation of sulfur(IV) by hydrogen peroxide, methylhydroperoxide and peroxyacetic acid. *J. Geophys. Res.* **92**, 4171-4177.

Martin L.R. (1984) Kinetic studies of sulfite oxidation in aqueous solution. In *SO_2, NO and NO_2 Oxidation Mechanisms: Atmospheric Considerations* (edited by Calvert J.G.), pp. 63-100, Butterworth Publishers, Boston.

Martin L.R., Damschen D.E. and Judeikis H.S. (1981) The reactions of nitrogen oxides with SO_2 in aqueous aerosols. *Atmospheric Environment* **15**, 191-195.

Monteith J.L. (1973) *Principles of Environmental Physics*, pp. 200-212, American Elsevier Publishing Company, Inc., New York.

Morel F.M.M. (1983) *Principles of aquatic chemistry*, pp. 314-328, John Wiley & Sons, New York.

Mudd J.B. (1975a) Sulfur dioxide. In *Responses of Plants to Air Pollution* (edited by Mudd J.B. and Kozlowski T.T.), pp. 9-22, Academic Press, Inc., New York.

Mudd J.B. (1975b) Peroxyacyl nitrates. In *Responses of Plants to Air Pollution* (edited by Mudd J.B. and Kozlowski T.T.), pp. 97-110, Academic Press, Inc., New York.

NCAR (1985) *The NCAR Eulerian Regional Acid Deposition Model.* NCAR/TN-256+STR, ADMP-85-3, National Center for Atmospheric Research, Boulder, CO, 202 pp.

NCAR (1986) *Preliminary Evaluation Studies with the Regional Acid Deposition Model (RADM).* NCAR/TN-256+STR, ADMP-86-4, National Center for Atmospheric Research, Boulder, CO, 202 pp.

Oblath S.B., Markowitz S.S., Novakov T. and Chang S.G. (1981) Kinetics of the formation of hydroxylamine disulfonate by reaction of nitrite with sulfites. *J. Phys. Chem.* **85**, 1017-1021.

Quinn J.A. and Otto N.C. (1971) Carbon dioxide exchange at the air-sea interface: Flux augmentation by chemical reaction. *J. Geophys. Res.* **76**, 1539-1549.

Schwartz S.E. (1984) Gas-aqueous reactions of sulfur and nitrogen oxides in liquid-water clouds. In *SO_2, NO, and NO_2 Oxidation Mechanisms: Atmospheric Considerations* (edited by Calvert J.G.), pp. 173-208, Butterworth Publishers, Boston.

Seinfeld J.H. (1986) *Atmospheric Chemistry and Physics of Air Pollution*, pp. 198-241, John Wiley & Sons, New York.

Sheih C.M., Wesely M.L. and Hicks B.B. (1979) Estimated dry deposition velocities of sulfur over the eastern United States and surrounding regions. *Atmospheric Environment* **13**, 1361-1368.

Sheih C.M., Wesely M.L. and Walcek C.J. (1986) *A Dry Deposition Module for Regional Acid Deposition.* EPA/600/3-86/037, U.S. Environmental Protection Agency (available as PB86218104 from NTIS, Springfield, VA) 63 pp.

Stumm W. and Morgan J.J. (1981) *Aquatic Chemistry*, pp. 437-459, John Wiley & Sons, New York.

Travis C.C. and Hattemer-Frey H.A. (1988) Uptake of organics by aerial plant parts: A call for research. *Chemosphere* 17, 277-283.

Unsworth M.H. (1980) The exchange of carbon dioxide and air pollutants between vegetation and the atmosphere. In *Plants and their Atmospheric Environment* (edited by Grace J., Ford E.D. and Jarvis P.G.), pp. 111-138. Blackwells, London.

van Hove L.W.A., Koops A.J., Adema E.H., Vredenberg W.J. and Pieters G.A. (1987) Analysis of the uptake of atmospheric ammonia by leaves of *Phaseolus vulgaris* L. *Atmospheric Environment* 21, 1759-1763.

Valdez M.P. (1987) Gaseous deposition of snow. 1. Experimental study of SO_2 and NO_2 Deposition. *J. Geophys. Res* 92, 9779-9787.

Walcek C.J., Brost R.A, Chang J.S. and Wesely M.L. (1986) SO_2, sulfate and HNO_3 deposition velocities computed using regional landuse and meteorological data. *Atmospheric Environment* 20, 949-964.

Wesely M.L. (1983) Turbulent transport of ozone to surfaces common in the eastern United States. In *Trace Atmospheric Constituents: Properties, Transformations, & Fates* (edited by Schwartz S.E.), pp. 345-370. John Wiley & Sons, Inc., New York.

Wesely M.L. and Hicks B.B. (1977) Some factors that affect the deposition rates of sulfur dioxide and similar gases on vegetation. *J. Air Pollut. Control Ass.*. 27, 1110-1116.

Wesely M.L., Cook D.R. and Hart R.L. (1983) Fluxes of gases and particles above a deciduous forest in wintertime. *Boundary-Layer Meteorol.* 90, 237-255.

Wesely M.L., Eastman J.A., Stedman D.H. and Yalvac E.D. (1982) An eddy-correlation measurement of NO_2 flux to vegetation and comparison to O_3 flux. *Atmospheric Environment* 16, 815-820.

Williams E.J., Parrish D.D. and Fehsenfeld F.C. (1987) Determination of nitrogen oxide emissions from soils: Results from a grassland site in Colorado, United States. *J. Geophys. Res.* 92, 2173-2179.

Appendix I. Computed surface resistances r_c (s m^{-1}).

Landuse type	Solar irradiation (W m^{-2})						
	800	500	300	100	0	0	0
						Wetted surface	
						dew	rain
Sulfur dioxide							
Seasonal category 1, midsummer with lush vegetation							
1	490.	490.	490.	490.	490.	40.	40.
2	80.	90.	100.	180.	290.	70.	300.
3	140.	140.	160.	250.	350.	80.	370.
4	120.	130.	160.	370.	1000.	90.	1100.
5	200.	210.	250.	500.	1000.	90.	1100.
6	160.	170.	200.	430.	940.	90.	1000.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	100.	100.	120.	190.	260.	70.	260.
10	120.	120.	140.	220.	300.	70.	310.
11	190.	200.	220.	360.	500.	80.	490.
Seasonal category 2, autumn with unharvested cropland							
1	490.	490.	490.	490.	490.	40.	40.
2	320.	320.	320.	320.	330.	70.	310.
3	400.	400.	400.	410.	410.	80.	390.
4	1300.	1300.	1300.	1300.	1400.	90.	1200.
5	380.	400.	460.	770.	1300.	90.	1300.
6	600.	620.	680.	960.	1300.	90.	1100.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	190.	190.	190.	190.	190.	60.	190.
10	380.	380.	380.	380.	390.	80.	370.
11	480.	480.	480.	480.	490.	80.	460.
Seasonal category 3, late autumn after frost, no snow							
1	490.	490.	490.	490.	490.	40.	40.
2	160.	160.	160.	160.	160.	160.	160.
3	410.	410.	410.	410.	420.	80.	390.
4	1100.	1100.	1100.	1100.	1100.	90.	1000.
5	740.	770.	820.	1000.	1300.	90.	1300.
6	940.	950.	990.	1100.	1200.	90.	1100.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	980.	980.	980.
9	100.	100.	100.	100.	100.	50.	90.
10	230.	230.	230.	230.	240.	70.	230.
11	460.	460.	460.	460.	480.	80.	450.
Seasonal category 4, winter with snow on ground							
1	210.	210.	210.	210.	210.	210.	210.
2	120.	120.	120.	120.	120.	120.	120.
3	120.	120.	120.	120.	120.	120.	120.
4	980.	980.	980.	990.	1000.	1000.	1000.
5	340.	380.	450.	680.	1300.	1300.	1300.
6	430.	460.	520.	690.	1200.	1200.	1200.
7	10.	10.	10.	10.	10.	10.	10.
8	990.	990.	990.	990.	990.	990.	990.
9	160.	160.	160.	160.	160.	160.	160.
10	120.	120.	120.	120.	120.	120.	120.
11	110.	110.	110.	110.	110.	110.	110.
Seasonal category 5, transitional spring with partial green coverage							
1	590.	590.	590.	590.	590.	40.	40.
2	110.	120.	120.	160.	180.	60.	180.
3	220.	230.	250.	320.	370.	80.	370.
4	260.	280.	330.	620.	1100.	90.	1000.
5	350.	370.	410.	640.	1000.	90.	1100.
6	310.	330.	370.	630.	1000.	90.	1000.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	120.	130.	140.	160.	180.	60.	180.
10	180.	180.	190.	240.	280.	70.	270.
11	290.	300.	320.	410.	470.	80.	450.

Appendix I continued

Ozone

Seasonal category 1, midsummer with lush vegetation

1	390.	390.	390.	390.	390.	390.	390.
2	70.	80.	90.	170.	290.	290.	240.
3	100.	110.	120.	180.	250.	250.	210.
4	100.	110.	130.	320.	950.	950.	580.
5	160.	170.	200.	420.	950.	950.	580.
6	130.	140.	170.	380.	970.	970.	590.
7	1900.	1900.	1900.	1900.	1900.	1900.	1900.
8	390.	390.	390.	390.	390.	390.	390.
9	110.	120.	140.	310.	790.	750.	500.
10	100.	100.	120.	190.	270.	270.	230.
11	130.	140.	160.	240.	350.	330.	270.

Seasonal category 2, autumn with unharvested cropland

1	390.	390.	390.	390.	390.	390.	390.
2	190.	200.	210.	240.	280.	260.	220.
3	190.	200.	210.	240.	280.	260.	220.
4	430.	460.	520.	700.	1200.	940.	570.
5	300.	320.	370.	630.	1200.	1000.	610.
6	360.	390.	440.	690.	1300.	1000.	610.
7	1900.	1900.	1900.	1900.	1900.	1900.	1900.
8	390.	390.	390.	390.	390.	390.	390.
9	360.	390.	420.	540.	820.	670.	460.
10	190.	200.	210.	240.	280.	260.	220.
11	210.	220.	230.	260.	310.	290.	240.

Seasonal category 3, late autumn after frost, no snow

1	390.	390.	390.	390.	390.	390.	390.
2	140.	140.	140.	150.	160.	160.	160.
3	190.	200.	210.	240.	280.	260.	220.
4	390.	420.	460.	600.	950.	760.	500.
5	500.	530.	570.	770.	1200.	1000.	610.
6	470.	500.	550.	740.	1200.	970.	590.
7	1900.	1900.	1900.	1900.	1900.	1900.	1900.
8	390.	390.	390.	390.	390.	390.	390.
9	500.	510.	540.	630.	890.	720.	490.
10	170.	180.	180.	190.	220.	210.	180.
11	220.	230.	230.	260.	300.	280.	230.

Seasonal category 4, winter with snow on ground

1	700.	700.	700.	700.	700.	700.	700.
2	900.	940.	1000.	1200.	2500.	2500.	2500.
3	900.	940.	1000.	1200.	2500.	2500.	2500.
4	550.	610.	700.	1000.	3000.	3000.	3000.
5	1000.	1000.	1100.	1300.	2300.	2300.	2300.
6	660.	700.	780.	1000.	2400.	2400.	2400.
7	1900.	1900.	1900.	1900.	1900.	1900.	1900.
8	410.	410.	410.	410.	410.	410.	410.
9	730.	770.	830.	1000.	2000.	2000.	2000.
10	820.	860.	910.	1100.	2000.	2000.	2000.
11	730.	770.	830.	1000.	2000.	2000.	2000.

Seasonal category 5, transitional spring with partial green coverage

1	390.	390.	390.	390.	390.	390.	390.
2	100.	100.	110.	150.	180.	180.	160.
3	140.	140.	160.	200.	250.	240.	210.
4	180.	190.	220.	430.	940.	810.	520.
5	300.	320.	360.	570.	900.	900.	580.
6	230.	240.	280.	500.	1000.	910.	560.
7	1900.	1900.	1900.	1900.	1900.	1900.	1900.
8	390.	390.	390.	390.	390.	390.	390.
9	190.	210.	240.	430.	850.	740.	490.
10	130.	130.	140.	180.	220.	210.	180.
11	170.	180.	190.	240.	290.	270.	230.

Appendix I continued

Nitrogen dioxide

Seasonal category 1, midsummer with lush vegetation

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	100.	100.	130.	370.	1400.	1400.	1200.
3	190.	200.	250.	620.	1700.	1600.	1500.
4	120.	130.	160.	480.	2800.	2700.	2300.
5	210.	230.	280.	770.	2800.	2700.	2300.
6	170.	180.	220.	660.	3300.	3100.	2600.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	140.	150.	180.	590.	5300.	4800.	3600.
10	160.	170.	210.	540.	1600.	1500.	1400.
11	230.	250.	300.	730.	1800.	1800.	1600.

Seasonal category 2, autumn with unharvested cropland

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	1100.	1100.	1100.	1200.	1400.	1300.	1200.
3	1300.	1300.	1300.	1400.	1700.	1600.	1500.
4	1800.	1800.	1800.	1900.	2600.	2400.	2100.
5	460.	500.	590.	1300.	3000.	2800.	2400.
6	780.	830.	970.	1700.	3400.	3100.	2600.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	2600.	2600.	2700.	2900.	4700.	4100.	3200.
10	1200.	1200.	1300.	1300.	1600.	1500.	1400.
11	1300.	1300.	1400.	1400.	1800.	1700.	1500.

Seasonal category 3, late autumn after frost, no snow

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	1300.	1300.	1300.	1300.	1400.	1400.	1400.
3	1300.	1300.	1400.	1400.	1700.	1600.	1500.
4	1700.	1700.	1700.	1800.	2300.	2200.	1900.
5	1200.	1200.	1400.	2100.	3000.	2800.	2400.
6	1600.	1600.	1800.	2300.	3300.	3000.	2500.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	4200.	4200.	4200.	4400.	5800.	4900.	3700.
10	1400.	1400.	1400.	1400.	1600.	1500.	1400.
11	1500.	1500.	1500.	1500.	1800.	1700.	1500.

Seasonal category 4, winter with snow on ground

1	5900.	5900.	5900.	5900.	5900.	5900.	5900.
2	7500.	7600.	7600.	7900.	9999.	9999.	9999.
3	7500.	7600.	7600.	7900.	9999.	9999.	9999.
4	3800.	3800.	3900.	4300.	9500.	9500.	9500.
5	8100.	8200.	8400.	9200.	9999.	9999.	9999.
6	4900.	5000.	5100.	5500.	9999.	9999.	9999.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	4000.	4000.	4000.	4000.	4000.	4000.	4000.
9	6000.	6100.	6100.	6400.	9900.	9900.	9900.
10	7000.	7000.	7100.	7400.	9999.	9999.	9999.
11	6000.	6100.	6100.	6400.	9900.	9900.	9900.

Seasonal category 5, transitional spring with partial green coverage

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	220.	240.	280.	640.	1300.	1300.	1200.
3	390.	420.	490.	940.	1700.	1600.	1500.
4	270.	290.	350.	850.	2400.	2300.	2000.
5	460.	500.	600.	1300.	2900.	2800.	2300.
6	360.	390.	470.	1100.	3100.	2900.	2400.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	320.	350.	430.	1100.	5300.	4700.	3600.
10	340.	370.	430.	870.	1600.	1500.	1400.
11	480.	510.	590.	1100.	1800.	1700.	1500.

Appendix I continued

Hydrogen peroxide

Seasonal category 1, midsummer with lush vegetation

1	260.	260.	260.	260.	260.	260.	260.
2	60.	60.	70.	130.	210.	60.	60.
3	80.	80.	90.	140.	180.	60.	60.
4	80.	90.	110.	250.	640.	80.	80.
5	130.	140.	160.	320.	640.	80.	80.
6	110.	120.	140.	290.	630.	80.	80.
7	0.	0.	0.	0.	0.	0.	0.
8	280.	280.	280.	280.	280.	280.	280.
9	70.	80.	90.	150.	230.	70.	60.
10	80.	80.	90.	140.	190.	60.	60.
11	110.	120.	130.	200.	270.	70.	70.

Seasonal category 2, autumn with unharvested cropland

1	260.	260.	260.	260.	260.	260.	260.
2	160.	160.	170.	190.	210.	60.	60.
3	150.	160.	170.	180.	210.	60.	60.
4	400.	430.	480.	640.	1000.	90.	80.
5	240.	260.	290.	500.	940.	90.	80.
6	310.	330.	380.	590.	1100.	90.	80.
7	0.	0.	0.	0.	0.	0.	0.
8	280.	280.	280.	280.	280.	280.	280.
9	140.	150.	150.	160.	180.	60.	60.
10	160.	160.	170.	190.	210.	60.	60.
11	180.	180.	190.	210.	250.	70.	60.

Seasonal category 3, late autumn after frost, no snow

1	260.	260.	260.	260.	260.	260.	260.
2	80.	80.	80.	80.	80.	80.	80.
3	160.	160.	170.	180.	210.	60.	60.
4	360.	380.	420.	540.	830.	80.	80.
5	400.	410.	450.	600.	940.	90.	80.
6	410.	430.	480.	640.	1000.	90.	80.
7	0.	0.	0.	0.	0.	0.	0.
8	280.	280.	280.	280.	280.	280.	280.
9	90.	90.	90.	90.	90.	40.	40.
10	120.	120.	120.	130.	130.	50.	50.
11	180.	180.	190.	200.	230.	60.	60.

Seasonal category 4, winter with snow on ground

1	190.	190.	190.	190.	190.	190.	190.
2	110.	110.	110.	110.	120.	120.	120.
3	110.	110.	110.	110.	120.	120.	120.
4	390.	420.	460.	610.	980.	980.	980.
5	300.	340.	400.	600.	1100.	1100.	1100.
6	330.	370.	420.	610.	1000.	1000.	1000.
7	10.	10.	10.	10.	10.	10.	10.
8	290.	290.	290.	290.	290.	290.	290.
9	130.	130.	140.	140.	150.	150.	150.
10	110.	110.	110.	110.	120.	120.	120.
11	100.	100.	100.	100.	110.	110.	110.

Seasonal category 5, transitional spring with partial green coverage

1	280.	280.	280.	280.	280.	280.	280.
2	70.	70.	90.	100.	110.	50.	50.
3	110.	110.	120.	150.	180.	60.	60.
4	150.	170.	190.	370.	740.	80.	80.
5	220.	230.	260.	410.	640.	80.	80.
6	190.	200.	230.	400.	720.	80.	80.
7	0.	0.	0.	0.	0.	0.	0.
8	280.	280.	280.	280.	280.	280.	280.
9	100.	100.	110.	140.	180.	60.	60.
10	90.	100.	100.	130.	150.	60.	50.
11	140.	140.	150.	190.	230.	60.	60.

Appendix I continued

Acetaldehyde

Seasonal category 1, midsummer with lush vegetation

1	9999.	9999.	9999.	9999.	9999.	9999.	9999.
2	300.	310.	340.	710.	9999.	9999.	9999.
3	410.	430.	490.	1200.	9999.	9999.	9999.
4	320.	330.	370.	790.	9999.	9999.	9999.
5	430.	450.	520.	1300.	9999.	9999.	9999.
6	380.	390.	440.	1000.	9999.	9999.	9999.
7	6400.	6400.	6400.	6400.	6400.	6400.	6400.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	320.	340.	370.	780.	6900.	6800.	6800.
10	380.	390.	440.	1000.	9999.	9999.	9999.
11	470.	490.	570.	1400.	9999.	9999.	9999.

Seasonal category 2, autumn with unharvested cropland

1	9999.	9999.	9999.	9999.	9999.	9999.	9999.
2	9999.	9999.	9999.	9999.	9999.	9999.	9999.
3	9999.	9999.	9999.	9999.	9999.	9999.	9999.
4	9999.	9999.	9999.	9999.	9999.	9999.	9999.
5	760.	810.	970.	2800.	9999.	9999.	9999.
6	1300.	1400.	1700.	5400.	9999.	9999.	9999.
7	6400.	6400.	6400.	6400.	6400.	6400.	6400.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	6500.	6500.	6500.	6500.	6500.	6500.	6500.
10	9999.	9999.	9999.	9999.	9999.	9999.	9999.
11	9999.	9999.	9999.	9999.	9999.	9999.	9999.

Seasonal category 3, late autumn after frost, no snow

1	9999.	9999.	9999.	9999.	9999.	9999.	9999.
2	9999.	9999.	9999.	9999.	9999.	9999.	9999.
3	9999.	9999.	9999.	9999.	9999.	9999.	9999.
4	9999.	9999.	9999.	9999.	9999.	9999.	9999.
5	2400.	2800.	3200.	9999.	9999.	9999.	9999.
6	4800.	5000.	6200.	9999.	9999.	9999.	9999.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	9999.	9999.	9999.	9999.	9999.	9999.	9999.
10	9999.	9999.	9999.	9999.	9999.	9999.	9999.
11	9999.	9999.	9999.	9999.	9999.	9999.	9999.

Seasonal category 4, winter with snow on ground

1	9999.	9999.	9999.	9999.	9999.	9999.	9999.
2	9999.	9999.	9999.	9999.	9999.	9999.	9999.
3	9999.	9999.	9999.	9999.	9999.	9999.	9999.
4	9999.	9999.	9999.	9999.	9999.	9999.	9999.
5	9999.	9999.	9999.	9999.	9999.	9999.	9999.
6	9999.	9999.	9999.	9999.	9999.	9999.	9999.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	9999.	9999.	9999.	9999.	9999.	9999.	9999.
10	9999.	9999.	9999.	9999.	9999.	9999.	9999.
11	9999.	9999.	9999.	9999.	9999.	9999.	9999.

Seasonal category 5, transitional spring with partial green coverage

1	9999.	9999.	9999.	9999.	9999.	9999.	9999.
2	470.	490.	580.	1400.	9999.	9999.	9999.
3	740.	790.	930.	2700.	9999.	9999.	9999.
4	510.	540.	630.	1600.	9999.	9999.	9999.
5	760.	810.	970.	2800.	9999.	9999.	9999.
6	630.	670.	780.	2200.	9999.	9999.	9999.
7	6400.	6400.	6400.	6400.	6400.	6400.	6400.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	520.	540.	620.	1400.	6800.	6700.	6700.
10	650.	690.	810.	2300.	9999.	9999.	9999.
11	880.	940.	1100.	3300.	9999.	9999.	9999.

Appendix I continued

Formaldehyde

Seasonal category 1, midsummer with lush vegetation

1	6300.	6300.	6300.	6300.	6300.	6300.	6300.
2	80.	90.	110.	350.	2300.	990.	990.
3	170.	180.	220.	690.	4500.	1200.	1200.
4	100.	110.	130.	450.	6800.	1300.	1300.
5	180.	200.	250.	790.	6800.	1300.	1300.
6	140.	150.	180.	560.	3000.	1100.	1100.
7	10.	10.	10.	10.	10.	10.	10.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	80.	90.	100.	200.	310.	260.	260.
10	140.	150.	180.	560.	3100.	1100.	1100.
11	210.	230.	280.	870.	5700.	1300.	1300.

Seasonal category 2, autumn with unharvested cropland

1	6300.	6300.	6300.	6300.	6300.	6300.	6300.
2	3200.	3200.	3200.	3200.	3200.	1100.	1100.
3	5200.	5200.	5200.	5200.	5200.	1200.	1200.
4	8100.	8100.	8100.	8100.	8100.	1300.	1300.
5	430.	470.	570.	1600.	7400.	1300.	1300.
6	710.	760.	890.	1800.	3100.	1000.	1000.
7	10.	10.	10.	10.	10.	10.	10.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	210.	210.	210.	210.	210.	190.	190.
10	4600.	4600.	4600.	4600.	4600.	1200.	1200.
11	5900.	5900.	5900.	5900.	5900.	1300.	1300.

Seasonal category 3, late autumn after frost, no snow

1	6400.	6400.	6400.	6400.	6400.	6400.	6400.
2	2400.	2400.	2400.	2400.	2400.	2400.	2400.
3	5300.	5300.	5300.	5300.	5300.	1200.	1200.
4	7800.	7800.	7800.	7800.	7800.	1300.	1300.
5	1400.	1500.	1800.	4000.	7800.	1300.	1300.
6	2000.	2100.	2300.	3500.	4500.	1200.	1200.
7	40.	40.	40.	40.	40.	40.	40.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	140.	140.	140.	140.	140.	130.	130.
10	3100.	3100.	3100.	3100.	3100.	1100.	1100.
11	5900.	5900.	5900.	5900.	5900.	1300.	1300.

Seasonal category 4, winter with snow on ground

1	2000.	2000.	2000.	2000.	2000.	2000.	2000.
2	1900.	1900.	1900.	1900.	1900.	1900.	1900.
3	1900.	1900.	1900.	1900.	1900.	1900.	1900.
4	2800.	2800.	2800.	2800.	2800.	2800.	2800.
5	1800.	1800.	1900.	2000.	2900.	2900.	2900.
6	2200.	2200.	2200.	2300.	2800.	2800.	2800.
7	300.	300.	300.	300.	300.	300.	300.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	1900.	1900.	1900.	1900.	1900.	1900.	1900.
10	1900.	1900.	1900.	1900.	1900.	1900.	1900.
11	1100.	1100.	1100.	1100.	1100.	1100.	1100.

Seasonal category 5, transitional spring with partial green coverage

1	7800.	7800.	7800.	7800.	7800.	7800.	7800.
2	200.	210.	260.	720.	2300.	990.	990.
3	400.	440.	530.	1400.	5000.	1200.	1200.
4	250.	270.	330.	1000.	7500.	1300.	1300.
5	430.	480.	570.	1600.	6600.	1300.	1300.
6	320.	350.	420.	1100.	4100.	1200.	1200.
7	10.	10.	10.	10.	10.	10.	10.
8	9999.	9999.	9999.	9999.	9999.	9999.	9999.
9	120.	120.	140.	180.	210.	190.	190.
10	330.	360.	440.	1100.	3700.	1100.	1100.
11	500.	550.	670.	1800.	6100.	1300.	1300.

Appendix I continued

Methyl hydroperoxide

Seasonal category 1, midsummer with lush vegetation

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	100.	100.	130.	370.	1400.	1300.	1200.
3	190.	200.	240.	620.	1700.	1600.	1400.
4	120.	130.	160.	480.	2800.	2500.	2100.
5	210.	230.	280.	770.	2800.	2500.	2100.
6	170.	180.	220.	650.	3200.	2800.	2400.
7	400.	400.	400.	400.	400.	400.	400.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	110.	120.	150.	330.	650.	640.	610.
10	160.	170.	210.	540.	1600.	1500.	1300.
11	230.	250.	300.	730.	1800.	1700.	1500.

Seasonal category 2, autumn with unharvested cropland

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	1100.	1100.	1100.	1200.	1400.	1300.	1200.
3	1300.	1300.	1300.	1400.	1700.	1600.	1400.
4	1800.	1800.	1800.	1900.	2600.	2300.	2000.
5	460.	500.	590.	1300.	3000.	2600.	2200.
6	780.	830.	960.	1700.	3300.	2800.	2300.
7	400.	400.	400.	400.	400.	400.	400.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	510.	510.	520.	520.	560.	550.	530.
10	1200.	1200.	1200.	1300.	1600.	1400.	1300.
11	1300.	1300.	1300.	1400.	1700.	1600.	1400.

Seasonal category 3, late autumn after frost, no snow

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	1200.	1200.	1200.	1300.	1300.	1300.	1300.
3	1300.	1300.	1300.	1400.	1700.	1600.	1400.
4	1700.	1700.	1700.	1800.	2300.	2100.	1800.
5	1200.	1200.	1400.	2100.	3000.	2600.	2200.
6	1600.	1600.	1700.	2300.	3300.	2800.	2300.
7	970.	970.	970.	970.	970.	970.	970.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	900.	900.	900.	910.	960.	910.	860.
10	1300.	1300.	1300.	1400.	1600.	1400.	1300.
11	1500.	1500.	1500.	1500.	1800.	1600.	1400.

Seasonal category 4, winter with snow on ground

1	5300.	5300.	5300.	5300.	5300.	5300.	5300.
2	6500.	6800.	6600.	6800.	9500.	9500.	9500.
3	6500.	6800.	6600.	6800.	9500.	9500.	9500.
4	3500.	3800.	3800.	4000.	8000.	8000.	8000.
5	6500.	6800.	6800.	7300.	9500.	9500.	9500.
6	4400.	4400.	4500.	4900.	8400.	8400.	8400.
7	5200.	5200.	5200.	5200.	5200.	5200.	5200.
8	4000.	4000.	4000.	4000.	4000.	4000.	4000.
9	5300.	5400.	5400.	5700.	8200.	8200.	8200.
10	6100.	6100.	6200.	6400.	8600.	8600.	8600.
11	4900.	5000.	5000.	5200.	7300.	7300.	7300.

Seasonal category 5, transitional spring with partial green coverage

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	220.	240.	280.	640.	1300.	1200.	1100.
3	390.	410.	480.	930.	1700.	1500.	1400.
4	270.	290.	350.	840.	2400.	2100.	1900.
5	460.	500.	600.	1300.	2900.	2600.	2200.
6	360.	390.	470.	1100.	3100.	2700.	2300.
7	400.	400.	400.	400.	400.	400.	400.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	210.	220.	250.	410.	570.	550.	530.
10	340.	360.	430.	860.	1500.	1400.	1300.
11	480.	510.	590.	1000.	1800.	1600.	1500.

Appendix I continued

Peroxyacetic acid

Seasonal category 1, midsummer with lush vegetation

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	120.	130.	160.	430.	1300.	1200.	1100.
3	230.	240.	290.	700.	1600.	1500.	1300.
4	140.	160.	190.	570.	2800.	2300.	2000.
5	260.	280.	340.	890.	2800.	2300.	2000.
6	200.	220.	270.	760.	3100.	2500.	2100.
7	180.	180.	180.	180.	180.	180.	180.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	120.	130.	150.	290.	460.	440.	430.
10	190.	210.	250.	610.	1500.	1400.	1200.
11	280.	300.	360.	820.	1800.	1600.	1400.

Seasonal category 2, autumn with unharvested cropland

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	1100.	1100.	1100.	1100.	1300.	1200.	1100.
3	1300.	1300.	1300.	1400.	1700.	1500.	1300.
4	1800.	1800.	1800.	1900.	2600.	2100.	1800.
5	650.	690.	700.	1400.	3000.	2400.	2100.
6	890.	940.	1000.	1800.	3200.	2500.	2100.
7	180.	180.	180.	180.	180.	180.	180.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	340.	340.	340.	350.	360.	350.	340.
10	1200.	1200.	1200.	1300.	1600.	1400.	1200.
11	1300.	1300.	1300.	1400.	1700.	1500.	1300.

Seasonal category 3, late autumn after frost, no snow

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	1200.	1200.	1200.	1200.	1300.	1300.	1300.
3	1300.	1300.	1300.	1400.	1700.	1500.	1300.
4	1700.	1700.	1700.	1800.	2300.	1900.	1700.
5	1300.	1400.	1500.	2200.	3000.	2400.	2100.
6	1700.	1700.	1800.	2300.	3200.	2500.	2100.
7	440.	440.	440.	440.	440.	440.	440.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	500.	500.	500.	500.	510.	490.	480.
10	1300.	1300.	1300.	1300.	1500.	1300.	1200.
11	1500.	1500.	1500.	1500.	1700.	1500.	1400.

Seasonal category 4, winter with snow on ground

1	4700.	4700.	4700.	4700.	4700.	4700.	4700.
2	5600.	5600.	5700.	5900.	7700.	7700.	7700.
3	5600.	5600.	5700.	5900.	7700.	7700.	7700.
4	3300.	3300.	3400.	3700.	6800.	6800.	6800.
5	5400.	5500.	5600.	5900.	7700.	7700.	7700.
6	3900.	4000.	4000.	4300.	7000.	7000.	7000.
7	2800.	2800.	2800.	2800.	2800.	2800.	2800.
8	3900.	3900.	3900.	3900.	3900.	3900.	3900.
9	4700.	4700.	4800.	5000.	6800.	6800.	6800.
10	5300.	5300.	5400.	5500.	7100.	7100.	7100.
11	4100.	4100.	4100.	4200.	5600.	5600.	5600.

Seasonal category 5, transitional spring with partial green coverage

1	2900.	2900.	2900.	2900.	2900.	2900.	2900.
2	260.	280.	330.	700.	1300.	1200.	1100.
3	450.	480.	550.	990.	1700.	1500.	1300.
4	320.	350.	420.	950.	2400.	2000.	1800.
5	580.	600.	700.	1400.	2800.	2400.	2000.
6	440.	470.	560.	1200.	3000.	2400.	2100.
7	180.	180.	180.	180.	180.	180.	180.
8	3800.	3800.	3800.	3800.	3800.	3800.	3800.
9	190.	200.	220.	300.	360.	350.	350.
10	400.	430.	490.	930.	1500.	1300.	1200.
11	550.	590.	670.	1100.	1800.	1500.	1400.

Appendix I continued

Formic acid

Seasonal category 1, midsummer with lush vegetation

1	100.	100.	100.	100.	100.	100.	100.
2	20.	20.	30.	30.	40.	0.	0.
3	20.	20.	20.	30.	30.	0.	0.
4	30.	30.	30.	40.	40.	0.	0.
5	30.	30.	30.	40.	40.	0.	0.
6	30.	30.	30.	40.	40.	0.	0.
7	0.	0.	0.	0.	0.	0.	0.
8	20.	20.	20.	20.	20.	20.	20.
9	30.	30.	30.	40.	50.	0.	0.
10	20.	20.	30.	30.	30.	0.	0.
11	40.	40.	50.	60.	60.	0.	0.

Seasonal category 2, autumn with unharvested cropland

1	100.	100.	100.	100.	100.	100.	100.
2	70.	70.	80.	80.	90.	0.	0.
3	60.	60.	60.	60.	70.	0.	0.
4	130.	140.	150.	160.	190.	0.	0.
5	60.	60.	70.	80.	90.	0.	0.
6	100.	110.	120.	150.	170.	0.	0.
7	0.	0.	0.	0.	0.	0.	0.
8	20.	20.	20.	20.	20.	20.	20.
9	80.	80.	90.	90.	100.	0.	0.
10	60.	70.	70.	70.	80.	0.	0.
11	70.	70.	70.	80.	80.	0.	0.

Seasonal category 3, late autumn after frost, no snow

1	100.	100.	100.	100.	100.	100.	100.
2	10.	10.	10.	10.	10.	10.	10.
3	60.	60.	60.	60.	70.	0.	0.
4	130.	130.	140.	160.	180.	0.	0.
5	70.	70.	70.	80.	90.	0.	0.
6	110.	120.	130.	150.	170.	0.	0.
7	0.	0.	0.	0.	0.	0.	0.
8	20.	20.	20.	20.	20.	20.	20.
9	60.	60.	60.	60.	60.	0.	0.
10	40.	40.	40.	40.	40.	0.	0.
11	70.	70.	70.	70.	80.	0.	0.

Seasonal category 4, winter with snow on ground

1	90.	90.	90.	90.	90.	90.	90.
2	10.	10.	10.	10.	10.	10.	10.
3	10.	10.	10.	10.	10.	10.	10.
4	270.	300.	330.	450.	660.	660.	660.
5	80.	90.	100.	120.	130.	130.	130.
6	100.	110.	130.	160.	190.	190.	190.
7	0.	0.	0.	0.	0.	0.	0.
8	20.	20.	20.	20.	20.	20.	20.
9	30.	40.	40.	40.	40.	40.	40.
10	10.	10.	10.	10.	10.	10.	10.
11	30.	30.	30.	40.	40.	40.	40.

Seasonal category 5, transitional spring with partial green coverage

1	100.	100.	100.	100.	100.	100.	100.
2	20.	20.	30.	30.	30.	0.	0.
3	30.	30.	40.	40.	40.	0.	0.
4	50.	60.	60.	80.	90.	0.	0.
5	30.	40.	40.	40.	40.	0.	0.
6	50.	50.	50.	60.	70.	0.	0.
7	0.	0.	0.	0.	0.	0.	0.
8	20.	20.	20.	20.	20.	20.	20.
9	40.	50.	50.	60.	60.	0.	0.
10	30.	30.	30.	30.	40.	0.	0.
11	60.	60.	60.	70.	70.	0.	0.

Appendix I continued

Ammonia

Seasonal category 1, midsummer with lush vegetation

1	2000.	2000.	2000.	2000.	2000.	2000.	2000.
2	60.	60.	80.	220.	830.	310.	310.
3	120.	130.	150.	420.	1400.	380.	380.
4	70.	80.	100.	310.	2600.	430.	430.
5	130.	140.	180.	520.	2600.	430.	430.
6	100.	110.	130.	390.	1800.	400.	400.
7	0.	0.	0.	0.	0.	0.	0.
8	4600.	4600.	4600.	4600.	4600.	4600.	4600.
9	60.	70.	80.	170.	290.	180.	180.
10	90.	100.	130.	340.	1000.	340.	340.
11	150.	160.	200.	540.	1800.	390.	390.

Seasonal category 2, autumn with unharvested cropland

1	2000.	2000.	2000.	2000.	2000.	2000.	2000.
2	1000.	1000.	1000.	1000.	1000.	340.	340.
3	1600.	1600.	1600.	1600.	1600.	380.	380.
4	3200.	3200.	3200.	3200.	3300.	430.	430.
5	300.	330.	390.	1000.	3100.	430.	430.
6	500.	530.	620.	1100.	1900.	400.	400.
7	0.	0.	0.	0.	0.	0.	0.
8	4600.	4600.	4600.	4600.	4600.	4600.	4600.
9	200.	200.	200.	200.	200.	140.	140.
10	1400.	1400.	1400.	1400.	1400.	370.	370.
11	1900.	1900.	1900.	1900.	1900.	390.	390.

Seasonal category 3, late autumn after frost, no snow

1	2000.	2000.	2000.	2000.	2000.	2000.	2000.
2	760.	760.	760.	760.	760.	760.	760.
3	1600.	1600.	1600.	1600.	1700.	380.	380.
4	2900.	2900.	2900.	2900.	2900.	430.	430.
5	930.	980.	1100.	2000.	3200.	430.	430.
6	1200.	1200.	1300.	1800.	2200.	410.	410.
7	10.	10.	10.	10.	10.	10.	10.
8	4600.	4600.	4600.	4600.	4600.	4600.	4600.
9	110.	110.	110.	110.	110.	90.	90.
10	1000.	1000.	1000.	1000.	1000.	330.	330.
11	1900.	1900.	1900.	1900.	1900.	390.	390.

Seasonal category 4, winter with snow on ground

1	680.	680.	680.	680.	680.	680.	680.
2	590.	590.	590.	590.	590.	590.	590.
3	590.	590.	590.	590.	590.	590.	590.
4	1500.	1500.	1500.	1500.	1500.	1500.	1500.
5	830.	860.	910.	1100.	1900.	1900.	1900.
6	1000.	1000.	1100.	1200.	1700.	1700.	1700.
7	90.	90.	90.	90.	90.	90.	90.
8	4700.	4700.	4700.	4700.	4700.	4700.	4700.
9	620.	620.	620.	620.	620.	620.	620.
10	580.	580.	580.	580.	580.	580.	580.
11	380.	380.	380.	380.	380.	380.	380.

Seasonal category 5, transitional spring with partial green coverage

1	2500.	2500.	2500.	2500.	2500.	2500.	2500.
2	130.	140.	170.	370.	750.	300.	300.
3	270.	290.	340.	770.	1500.	380.	380.
4	190.	190.	230.	670.	2800.	430.	430.
5	300.	320.	390.	960.	2600.	430.	430.
6	220.	240.	290.	740.	1900.	400.	400.
7	0.	0.	0.	0.	0.	0.	0.
8	4600.	4600.	4600.	4600.	4600.	4600.	4600.
9	100.	100.	120.	160.	200.	140.	140.
10	220.	230.	280.	610.	1100.	350.	350.
11	330.	360.	430.	960.	1900.	400.	400.

Appendix I continued

Peroxyacetyl nitrate

Seasonal category 1, midsummer with lush vegetation

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	150.	160.	200.	510.	1400.	1400.	1200.
3	290.	310.	370.	820.	1700.	1600.	1500.
4	190.	200.	250.	700.	2800.	2700.	2300.
5	330.	350.	430.	1000.	2800.	2700.	2300.
6	260.	280.	350.	940.	3300.	3100.	2600.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3900.	3900.	3900.	3900.	3900.	3900.	3900.
9	220.	240.	290.	850.	4500.	4100.	3200.
10	240.	260.	310.	720.	1600.	1500.	1400.
11	350.	370.	440.	950.	1800.	1800.	1600.

Seasonal category 2, autumn with unharvested cropland

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	1100.	1100.	1100.	1200.	1400.	1300.	1200.
3	1300.	1300.	1300.	1400.	1700.	1600.	1500.
4	1800.	1800.	1800.	1900.	2800.	2400.	2100.
5	680.	730.	850.	1600.	3000.	2800.	2300.
6	1000.	1100.	1200.	2000.	3400.	3100.	2600.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3900.	3900.	3900.	3900.	3900.	3900.	3900.
9	2400.	2400.	2500.	2600.	4100.	3600.	2900.
10	1200.	1300.	1300.	1300.	1600.	1500.	1400.
11	1300.	1300.	1400.	1400.	1800.	1700.	1500.

Seasonal category 3, late autumn after frost, no snow

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	1300.	1300.	1300.	1300.	1400.	1400.	1400.
3	1300.	1300.	1400.	1400.	1700.	1700.	1500.
4	1700.	1700.	1700.	1800.	2400.	2200.	1900.
5	1500.	1600.	1700.	2300.	3000.	2800.	2400.
6	1800.	1900.	2000.	2400.	3300.	3000.	2500.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3900.	3900.	3900.	3900.	3900.	3900.	3900.
9	4000.	4000.	4000.	4100.	5400.	4600.	3500.
10	1400.	1400.	1400.	1400.	1600.	1500.	1400.
11	1500.	1500.	1500.	1600.	1800.	1700.	1500.

Seasonal category 4, winter with snow on ground

1	6000.	6000.	6000.	6000.	6000.	6000.	6000.
2	7700.	7700.	7800.	8100.	9999.	9999.	9999.
3	7700.	7700.	7800.	8100.	9999.	9999.	9999.
4	3800.	3900.	4000.	4400.	9700.	9700.	9700.
5	8400.	8500.	8700.	9200.	9999.	9999.	9999.
6	5000.	5000.	5100.	5500.	9900.	9900.	9900.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	4000.	4000.	4000.	4000.	4000.	4000.	4000.
9	6100.	6100.	6200.	6500.	9999.	9999.	9999.
10	7100.	7100.	7200.	7500.	9999.	9999.	9999.
11	6100.	6100.	6200.	6500.	9999.	9999.	9999.

Seasonal category 5, transitional spring with partial green coverage

1	3000.	3000.	3000.	3000.	3000.	3000.	3000.
2	330.	350.	410.	800.	1300.	1300.	1200.
3	540.	570.	650.	1000.	1700.	1600.	1500.
4	400.	430.	510.	1000.	2400.	2300.	2000.
5	680.	730.	850.	1600.	2900.	2800.	2300.
6	540.	580.	690.	1400.	3100.	2900.	2400.
7	9999.	9999.	9999.	9999.	9999.	9999.	9999.
8	3900.	3900.	3900.	3900.	3900.	3900.	3900.
9	490.	530.	640.	1500.	4500.	4000.	3200.
10	490.	510.	590.	1000.	1600.	1500.	1400.
11	660.	690.	780.	1200.	1800.	1700.	1500.

Appendix I continued

Nitrous acid

Seasonal category 1, midsummer with lush vegetation

1	490.	490.	490.	490.	490.	490.	490.
2	70.	80.	90.	170.	290.	70.	70.
3	120.	130.	150.	240.	350.	80.	80.
4	100.	110.	140.	340.	1000.	90.	90.
5	170.	190.	220.	460.	1000.	90.	90.
6	140.	150.	180.	400.	940.	90.	90.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	90.	90.	100.	180.	260.	70.	70.
10	100.	110.	130.	210.	300.	70.	70.
11	170.	180.	200.	340.	500.	80.	80.

Seasonal category 2, autumn with unharvested cropland

1	490.	490.	490.	490.	490.	490.	490.
2	320.	320.	320.	320.	330.	70.	70.
3	400.	400.	400.	410.	410.	80.	80.
4	1300.	1300.	1300.	1300.	1400.	90.	90.
5	340.	370.	420.	730.	1300.	90.	90.
6	550.	580.	640.	930.	1300.	90.	90.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	190.	190.	190.	190.	190.	60.	60.
10	380.	380.	380.	380.	390.	80.	80.
11	480.	480.	480.	480.	490.	80.	80.

Seasonal category 3, late autumn after frost, no snow

1	490.	490.	490.	490.	490.	490.	490.
2	160.	160.	160.	160.	160.	160.	160.
3	410.	410.	410.	410.	420.	80.	80.
4	1100.	1100.	1100.	1100.	1100.	90.	90.
5	710.	730.	780.	1000.	1300.	90.	90.
6	910.	920.	960.	1100.	1200.	90.	90.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	100.	100.	100.	100.	100.	50.	50.
10	230.	230.	230.	230.	240.	70.	70.
11	460.	460.	460.	460.	480.	80.	80.

Seasonal category 4, winter with snow on ground

1	210.	210.	210.	210.	210.	210.	210.
2	120.	120.	120.	120.	120.	120.	120.
3	120.	120.	120.	120.	120.	120.	120.
4	980.	980.	980.	990.	1000.	1000.	1000.
5	340.	380.	450.	680.	1300.	1300.	1300.
6	430.	460.	520.	690.	1200.	1200.	1200.
7	10.	10.	10.	10.	10.	10.	10.
8	990.	990.	990.	990.	990.	990.	990.
9	160.	160.	160.	160.	160.	160.	160.
10	120.	120.	120.	120.	120.	120.	120.
11	110.	110.	110.	110.	110.	110.	110.

Seasonal category 5, transitional spring with partial green coverage

1	590.	590.	590.	590.	590.	590.	590.
2	100.	110.	120.	160.	180.	60.	60.
3	210.	220.	240.	310.	370.	80.	80.
4	230.	250.	290.	580.	1100.	90.	90.
5	320.	330.	380.	620.	1000.	90.	90.
6	280.	290.	340.	600.	1000.	90.	90.
7	0.	0.	0.	0.	0.	0.	0.
8	970.	970.	970.	970.	970.	970.	970.
9	120.	120.	130.	160.	180.	60.	60.
10	160.	170.	180.	240.	280.	70.	70.
11	270.	280.	300.	400.	470.	80.	80.

Appendix II. Code for the Argonne dry deposition module.

```
C .   MODULE.FOR
C*** Argonne National Laboratory version of a dry deposition
C     module that can be adapted for use in RADM.
C*** This module is an uptdated version, as of 3/1/88,
C     and contains elements of the version completed on 6/11/86.
C*** This revision was made by M. L. Wesely and B. M. Lesht,
C     and is now designed to computed dry deposition velocities
C     of a greater number of substances, and with a more explicit
C     derivation of surface resistances.
C*** The main program and the subroutine METERO show below are
C     used only as an example for the use of the deposition
C     velocity subroutine DEPVEL.
C
C*** The following main program substitutes inputs from an
C     atmospheric transport model (e.g., RADM).
C
      IMPLICIT INTEGER*2 (I-N)
      LOGICAL*1 COM(70),DUMMY(70)
      COMMON/METVAR/ HV(328,186),UUSTAR(328,186),
     1 RADIAT(328,186),TCZ(328,186),TC0(328,186),
     2 ZINV(328,186),USTAR(328,186),VD(15,328,186)
C
C*** Open file for values of maximum SO4 deposition velocities.
C     (VDSMAX), surface roughness lengths (IZO), and surface
C     components (RI, RLU, RAC, RGSS, RGSO, RCLS, RCLO).
        OPEN(UNIT=5,FILE='MODINP.DAT',RECORDTYPE='VARIABLE',
     1STATUS='OLD',FORM='FORMATTED')
C
C*** Open direct access file for landuse map.
        OPEN(UNIT=10,FILE='$DISK1:[LESHT.LANDUSE]DNEM085.DAT',
     1ACCESS='DIRECT',READONLY,RECL=14,STATUS='OLD')
C
C*** Open file for optional, averaged output for test runs.
      OPEN(UNIT=8,FILE='MODAV.DAT',RECORDTYPE='VARIABLE',
     1 STATUS='NEW',FORM='FORMATTED')
C
C*** Define season and height of required deposition velocity.
      INIT=1
      CZ=10.
      DO 10 ISESN=1,5
C
C*** Call METERO to obtain meteorological parameters for
C     computation of deposition velocity.
      CALL METERO(ISESN,IDEW,IRAIN)
      CALL DEPVEL(INIT,CZ,ISESN,IDEW,IRAIN)
   10 CONTINUE
C
C*** INIT=1 defined intially in data assignment to permit
C     initialization of parameters only once and then set
C     INIT=0 to bypass initialization in subroutine DEPVEL.
C
      INIT=0
      STOP
      END
```

```
      SUBROUTINE METERO(ISESN,IDEW,IRAIN)
C*****************************************************
C*** Meteorological module
      IMPLICIT INTEGER*2 (I-N)
      DIMENSION SOLAR(5),TEMP(5)
      COMMON/METVAR/ HV(328,186),UUSTAR(328,186),
     1 RADIAT(328,186),TCZ(328,186),TC0(328,186),
     2 ZINV(328,186),USTAR(328,186),VD(15,328,186)
C*** TEMP is the surface temperature (C) assumed for preliminary
C    output, here specific to season.
      DATA TEMP/25.,10.,2.,0.,10./
C*** SOLAR is the insolation (W/m**2) assumed for preliminary
C    output; here we can choose any level.
      DATA SOLAR/800.,500.,300.,100.,0./
      II=2
C*** Set IDEW = 1 for dewfall, IRAIN = 1 for rain-wetted surface.
      IDEW=0
      IRAIN=0
C*** Initialize variables for all locations.
      LONOB=328
      LATOB=186
      CU=3.9
      DO 10 I=1,LONOB
      DO 10 J=1,LATOB
C*** These dummy values are only to provide a test case.
      HV(I,J)=150.
      USTAR(I,J)=0.30
      UUSTAR(I,J)=CU*USTAR(I,J)
      RADIAT(I,J)=SOLAR(II)
      TCZ(I,J)=TEMP(ISESN)
      TC0(I,J)=TEMP(ISESN)
      ZINV(I,J)=1000.
   10 CONTINUE
      RETURN
      END
```

```
      SUBROUTINE DEPVEL(INIT,CZ,ISESN,IDEW,IRAIN)
C*************************************************************************
C
C*** This subroutine was constructed by M. L. Wesely and
C    B. M. Lesht, with earlier imput by C. M. Sheih, at
C    Argonne National laboratory.
C*** This subroutine is for computing the dry deposition
C    velocities for for (1) SO2, (2) O3, (3) NO2, (4) NO,
C    (5) HNO3, (6) H2O2, (7) ALD actually acetaldehyde,
C    (8) HCHO, (9) OP actually methyl hydroperoxide, (10) PAA,
C    (11) ORA actually formic acid, (12) NH3, (13) PAN, (14) HNO2,
C    and (15) SO4.
C*** No estimates are made of surface emissions.
C
C*** Input parameters are ISESN, ---, and ZINV as defined below:
C    INIT= 1 (yes) or 0 (no) for initialization of parameters;
C    ISESN = 1, midsummer with lush vegetation
C            2, autumn with unharvested cropland
C            3, late autumn after frost, no snow
C            4, winter with snow on ground
C            5, transitional spring with partial green coverage;
C    HV = virtual heat flux (W/m**2);
C    USTAR = friction velocity (m/s);
C    RADIAT = solar irradiation (W/m**2);
C    CZ = height (m) where deposition velocities are needed;
C    TCZ = temperature at height CZ;
C    TC0 = surface temperature;
C    ZINV = inversion height (m).
C*** Some other code names are as follows:
C    VDSMAX = maximum permissable surface SO4 deposition
C    velocity for various landuse types and seasons;
C    HSTAR = Henry's Law constant for substances 1 through 14,
C            for water pH near 7;
C    F0 = measure of reactivity for substances 1 through 14,
C         relative to O3;
C    DIFRAT = ratio of molecular diffusivity for water vapor
C             in air to diffusivity for substances 1 through 14;
C    VD = deposition velocities (m/s);
C    R = bulk surface (non-gaseous) resistances.
C
C*** Resistances and deposition velocities are computed for
C    specific landuse types in the landuse map, which was
C    derived from surface landuse and land cover inventory
C    by S. H. Page of Lockheed Engineering and Management
C    Services Inc., Remote Sensing Laboratory, Las Vegas,
C    Nevada 89114.
C*** There are eleven landuse types as follows:
C     1, urban land, almost no vegetation;
C     2, agricultural land, usually well watered;
C     3, range land, usually with low soil moisture;
C     4, deciduous forest;
C     5, coniferous forest;
C     6, mixed forest including wetland;
C     7, water, sea water assumed but used for fresh water also;
C     8, barren land, mostly desert;
C     9, non-forested wetland;
C    10, mixed agricultrual and range land;
C    11, rocky open areas occupied by low growing schrubs.
C*** The landuse data set is arranged in latitude degree,
C    number of 1/12 lat. deg. (which is at the grid center,
C    with grid increments of 1/6 deg), longitude degree,
C    number of 1/8 long. deg. (with increments of 1/4 deg.),
C    and the 11 values of percentage cover of each landuse
C    type.
C*** The grid domain is from 52+1/8 to 133+7/8 deg. west longitude
C    and from 24+1/12 to 54+11/12 deg. north latitude, for which
C    there are NX=328 points and NY=186 points.  Grid points start
C    from the SE corner and runs from E to W and from S to N.
C
C
```

```
C*** Set variables for direct access file for landuse map.
      IMPLICIT INTEGER*2 (I-N)
      INTEGER*4 NREC,IROW,ICOL
C
      LOGICAL*1 COM(70),DUMMY(70)
      DIMENSION IUSE(11),HSTAR(14),F0(14),DIFRAT(14),
     1 IZO(11),ZO(11),CXO(328),CYO(186),VDSMAX(11),
     2 XRI(11),XRLU(11),XRAC(11),XRGSS(11),XRGSO(11),XRCLS(11),
     3 XRCLO(11),RI(11),RLU(11),RAC(11),RGSS(11),RGSO(11),RCLS(11),
     4 RCLO(11),R(15,11),DC(14),C1X(14),ITUSE(11),IPUSE(11),VX(15)
      COMMON/METVAR/ HV(328,186),UUSTAR(328,186),
     1 RADIAT(328,186),TCZ(328,186),TC0(328,186),
     2 ZINV(328,186),USTAR(328,186),VD(15,328,186)
C
C*** Set values for substance chemical and physical properties.
      DATA HSTAR/1.0E+5,0.01,.01,.003,1.0E+14,1.0E+05,15.,6000.,240.
     1,540.,4.0E+06,20000.,3.6,1.0E+05/
      DATA F0/0.,1.,.1,0.,0.,1.,0.,0.,.1,.1,0.,0.,.1,0./
      DATA DIFRAT/1.9,1.6,1.6,1.3,1.9,1.4,1.6,1.3,1.6,2.0,1.6,.97,
     1 2.6,1.6/
C
C     THETA is the terrain slope, assumed to be zero radians for RADM
      THETA=0.
C
C
C*** READ IN MODINP.DAT
      IF(INIT.NE.1) GOTO 25
      PRINT *,' Reading in inputs for SO4 max dep. vel., Zo,
     1 and resistance components'
      LONST=1
      LATST=1
      LONOB=328
      LATOB=186
C*** Use the following four values to only consider 5X5 deg.
C     square centered at 92.5 deg. long. and 42.5 deg. lat.
C       LONST=153
C       LONOB=173
C       LATST=97
C       LATOB=127
C*** Set up coordinates (if needed) for the grid network.
C       C1=52.-1./8.
C       DO 10 I=1,LONOB
C    10 CXO(I)=C1+I/4.
C       C1=24.-1./12.
C       DO 12 J=1,LATOB
C    12 CYO(J)=C1+J/6.
C*** Read in maximum permissible SO4 deposition velocities.
C*** I is landuse type.
C*** Read a dummy line first.
      READ(5,1000) DUMMY
      READ(5,1000) DUMMY
 1000 FORMAT(70A1)
      DO 16 L=1,5
      IF(L.NE.ISESN) GOTO 14
      READ(5,1100) COM,(VDSMAX(I),I=1,11)
 1100 FORMAT(70A1/(11F8.3))
      GOTO 16
   14 READ(5,1100) COM,(C1,I=1,11)
   16 CONTINUE
C*** Read a dummy line.
      READ(5,1000) DUMMY
C*** Read IZO (surface roughness) of proper season.
      DO 22 L=1,5
      IF(L.NE.ISESN) GOTO 20
      READ(5,1200) COM,(IZO(I),I=1,11)
 1200 FORMAT(70A1/11I8)
      GOTO 22
   20 READ(5,1200) COM,(IC1,I=1,11)
   22 CONTINUE
C*** Read a dummy line.
      READ(5,1000) DUMMY
```

```
C*** Read in inital values of surface resistance components.
      L=0
   23 L=L+1
      READ(5,1000) COM
      READ(5,1400) (XRI(I),I=1,11)
      READ(5,1400) (XRLU(I),I=1,11)
      READ(5,1400) (XRAC(I),I=1,11)
      READ(5,1400) (XRGSS(I),I=1,11)
      READ(5,1400) (XRGSO(I),I=1,11)
      READ(5,1400) (XRCLS(I),I=1,11)
      READ(5,1400) (XRCLO(I),I=1,11)
 1400 FORMAT(5X,11F6.0)
      IF(L.NE.ISESN) GOTO 23
      REWIND 5
C*** Convert to appropriate units.
      DO 24 I=1,11
      ZO(I)=IZO(I)*1.E-4
   24 CONTINUE
   25 CONTINUE
C
C*** This is the end of parameter initialization and
C    the beginning of computing deposition velocities.
C
C
C*** BEGIN ITERATING THROUGH GEOGRAPHICAL DOMAIN.
C*** Flag for subroutine AVERAGE.
      IFLAG=1
C*** Step through all grid points.
      PRINT *,' Beginning iterations for seasonal
     1 category number ',ISESN
      DO 70 J=LATST,LATOB
      DO 70 I=LONST,LONOB
C*** Initialize variables.
      C1=(273.+TC0(I,J))/273.
      C2=C1**1.75*1.0E-04
      DO 31 K=1,15
      VD(K,I,J)=0.
   31 DC(K)=0.219*C2/DIFRAT(K)
      XNU=0.151*C1**1.77*1.0E-04
C
C*** Compute bulk surface resistances for gases.
C*** Adjust external surface resistances for temperature.
      RT=1000.*EXP(-TC0(I,J)-4.)
      DO 40 N=1,11
      RI(N)=XRI(N)
      RLU(N)=XRLU(N)+RT
      RAC(N)=XRAC(N)
      RGSS(N)=XRGSS(N)+RT
      RGSO(N)=XRGSO(N)+RT
      RCLS(N)=XRCLS(N)+RT
   40 RCLO(N)=XRCLO(N)+RT
      DO 44 N=1,11
C*** Set initial max. and min. values for resistance components.
      IF(RI(N).GE.9999.) RI(N)=100000.
      IF(RLU(N).GE.9999.) RLU(N)=100000.
      IF(RAC(N).GE.9999.) RAC(N)=100000.
      IF(RGSS(N).GE.9999.) RGSS(N)=100000.
      IF(RGSO(N).GE.9999.) RGSO(N)=100000.
      IF(RCLS(N).GE.9999.) RCLS(N)=100000.
      IF(RCLO(N).GE.9999.) RCLO(N)=100000.
      IF(RAC(N).LT.1.) RAC(N)=1.
      IF(RGSS(N).LT.1.) RGSS(N)=1.
      GFACTOR=100.
      RIX=RI(N)
C*** Adjust stomatal resistances for insolation and temp.
      IF(RI(N).GE.9999.) GOTO 42
      IF(TC0(I,J).LE.0..OR.TC0(I,J).GE.40.) GOTO 41
      GFACTOR=400./TC0(I,J)/(40.-TC0(I,J))
   41 RIX=RI(N)*(1.+(200./(RADIAT(I,J)+0.1))**2.)*GFACTOR
C*** Adjust stomatal resistances for liquid water coverage.
      IF(IDEW.EQ.1.OR.IRAIN.EQ.1) RIX=3.*RIX
```

```
C*** Compute aerodynamic resistance to lower elements in lower
C     part of canopy or structure.
   42 RDC=100.*(1.+1000./(RADIAT(I,J)+10.))/(1.+1000.*THETA)
C*** Compute resistances for SO2.
      X=1./(3.*RLU(N))
      RLUX=RLU(N)
      IF(TC0(I,J).GT.0.5) THEN
C     Adjust for effects of dew and rain.
      IF(RLUX.LT.9999..AND.IDEW.EQ.1) RLUX=100.
      IF(RLUX.LT.9999..AND.IRAIN.EQ.1) RLUX=1./(1./5000.+X)
      IF(N.EQ.1.AND.(IDEW+IRAIN).GE.1) RLUX=50.
      ENDIF
      R(1,N)=1./(1./(DIFRAT(1)*RIX)+1./RLUX+1./(RAC(N)+RGSS(N))
     1 +1./(RDC+RCLS(N)))
C*** Compute resistance for O3.
      RLUX=RLU(N)
      IF(TC0(I,J).GT.0.5) THEN
C     Adjust for effects of dew and rain.
      IF(RLUX.LT.9999..AND.IDEW.EQ.1) RLUX=1./(1./3000.+X)
      IF(RLUX.LT.9999..AND.IRAIN.EQ.1) RLUX=1./(1./1000.+X)
      ENDIF
      R(2,N)=1./(1./(DIFRAT(2)*RIX)+1./RLUX+1./(RAC(N)+RGS0(N))
     1 +1./(RDC+RCL0(N)))
C*** Compute resistances for (3) NO2,(4) NO, (5) HNO3, AND (4-12)
C     other substances for which solubility, aqueous redox, and
C     diffusivity ratio relative to water vapor in air are specified.
      DO 43 K=3,14
      RIXX=RIX*DIFRAT(K)+1./(HSTAR(K)/3000.+100*F0(K))
      RLUXX=RLU(N)/(HSTAR(K)/1.0E+5 + F0(K))
C     Adjust for effects of dew or rain
      IF((IDEW+IRAIN).GE.1.AND.TC0(I,J).GT.0.5.AND.RLU(N).LT.9999.)
     1 RLUXX=1./(1./(3.*RLUXX)+HSTAR(K)/1.0E+07+F0(K)/RLUX)
      RGSX=1./(HSTAR(K)/1.0E+05/RGSS(N)+F0(K)/RGS0(N))
      RCLX=1./(HSTAR(K)/1.0E+05/RCLS(N)+F0(K)/RCL0(N))
      R(K,N)=1./(1./RIXX+1./RLUXX+1./(RAC(N)+RGSX)
     1 +1./(RDC+RCLX))
   43 CONTINUE
   44 CONTINUE
C*** Set max. and min. values for bulk surface resistances.
      DO 45 K=1,14
      DO 45 N=1,11
      IF(R(K,N).GT.9999.)R(K,N)=9999.
   45 IF(R(K,N).LE.1.)R(K,N)=10.
C
C*** Read landuse data.
      ICOL=I
      IROW=J
      NREC=(IROW-1)*328+ICOL
      DO 46 IL=1,11
   46 IUSE(IL)=0.
      READ(10'NREC) LAT,LATF,LON,LONF,NPAIR,
     1 (ITUSE(IL),IPUSE(IL),IL=1,NPAIR)
      DO 47 IL=1,NPAIR
   47 IUSE(ITUSE(IL))=IPUSE(IL)
C
C*** Loop through different landuse types.
      DO 70 N=1,11
      IF(IUSE(N).EQ.0) GOTO 70
C*** Transfer heat flux to CHV to avoid being erased.
      CHV=HV(I,J)
      IF(N.NE.7) GOTO 50
C*** Set values of roughness length and heat flux for water.
      Z0(7)=1.4E-02*USTAR(I,J)*USTAR(I,J)/9.8
     1  +1.1E-01*XNU/USTAR(I,J)
      CHV=20.
   50 CONTINUE
```

```
C*** Compute local USTAR by assumming U*USTAR (UUSTAR) constant.
C     First choice assumes neutral conditions.
      CUSTA=SQRT(0.4*UUSTAR(I,J)/(ALOG(CZ/Z0(N))))
      ICNT=0
C     Start iteration.
   60 XX=CUSTA
      C1=-0.00327*CZ*CHV/((273.+TCZ(I,J))*USTAR(I,J)**3.)
      IF(C1.LT.0.) GOTO 61
      IF(C1.GE.1.) C1=0.99
      SIM=-5.*C1
      GOTO 62
   61 IF(C1.LE.-1.) C1=-0.99
      C2=ALOG(-C1)
      SIM=EXP(0.032+0.448*C2-0.132*C2*C2)
   62 CONTINUE
      IF(N.EQ.7) Z0(7)=1.4E-02*USTAR(I,J)*USTAR(I,J)/
     1  9.8+1.1E-01*XNU/USTAR(I,J)
      CUSTA=SQRT(0.4*UUSTAR(I,J)/(ALOG(CZ/Z0(N))-SIM))
      ERR=ABS((CUSTA-XX)/XX)
      ICNT=ICNT+1
      IF(ICNT.GT.5) GOTO 63
      IF(ERR.GT.0.01) GOTO 60
   63 SIC=SIM
      IF(C1.LT.0.) SIC=EXP(0.598+0.39*C2-0.09*C2*C2)
      CKUSTA=0.4*CUSTA
      OBKHOV=CZ/C1
C*** Compute SO4 surface deposition velocities.
      VDS=0.002*USTAR(I,J)
      IF(OBKHOV.LT.0.) VDS=VDS*(1.+(-300./OBKHOV)**0.6667)
      C1=ZINV(I,J)/OBKHOV
      IF(C1.LT.-30.) VDS=0.0009*USTAR(I,J)*(-C1)**0.6667
C*** Set VDS to be less than VDSMAX.
      CVDS=VDS
      IF(CVDS.GT.VDSMAX(N)) CVDS=VDSMAX(N)
      IF(Z0(N).LE.0.001) GOTO 65
      C1=ALOG(CZ/Z0(N))-SIC
      DO 64 K=1,14
C*** Compute deposition velocities for each grid point.
   64 C1X(K)=C1+CKUSTA*R(K,N)+1.75*(DIFRAT(K))**0.667
      GOTO 67
   65 CONTINUE
      DO 66 K=1,14
   66 C1X(K)=ALOG(CZ*CKUSTA/DC(K))+CKUSTA*R(K,N)-SIC
   67 DO 68 K=1,14
      VX(K)=VD(K,I,J)
   68 VD(K,I,J)=CKUSTA*IUSE(N)/C1X(K)
      VX(15)=VD(15,I,J)
      VD(15,I,J)=CKUSTA*IUSE(N)/(ALOG(CZ/Z0(N))
     1  +CKUSTA/CVDS-SIC)
C*** Optional averaging routine to get values of deposition
C     velocities, etc., for 5 by 5 deg increments.
      IF(I.EQ.LONOB.AND.J.EQ.LATOB) IFLAG=2
      CALL AVERAGE(I,J,N,ISESN,IUSE,USTAR,R,CVDS,VD,IFLAG)
      DO 69 K=1,15
   69 VD(K,I,J)=VX(K)+VD(K,I,J)
   70 CONTINUE
C
C
C*** THIS ENDS MAIN COMPUTATIONAL LOOP.
      PRINT *,' End of computations for seasonal
     1 category number ',ISESN
C*** Multiplication by 0.01 because IUSE(LL) was in %.
      DO 81 J=1,LATOB
      DO 81 I=1,LONOB
      DO 80 K=1,14
C*** The VD's comprise the primary output of this module. ***
   80 VD(K,I,J)=0.01*VD(K,I,J)
   81 VD(15,I,J)=0.01*VD(15,I,J)
      RETURN
      END
```

```
      SUBROUTINE AVERAGE(I,J,LL,ISESN,IUSE,USTAR,R,CVDS,VD,IFLAG)
C*************************************************************
C*** This subroutine is optional for checking out the module
C    for a limited amount of input data.  It is not intended for
C    use with RADM or other numerical models.  By changing when
C    IFLAG is set to 2 in the calling program, averages over
C    long assumed time periods may be computed.  Then, thAe
C    numerical value of 600 used between statements 30 and 40 of
C    this subroutine would have to be changed to the actual
C    number of samples used in the summations.
C*** Four matrices are produced whenever averages are produced
C    (IFLAG=2, which routes program to statement label 30).
C    The matrices consist of (1) a compressed landuse file with
C    USTAR values for each 5 by 5 deg square, and (2-4) values
C    of bulk surface resistances for 15 substances.
C
      IMPLICIT INTEGER*2 (I-N)
      INTEGER*4 IUSEA
      DIMENSION IUSE(11),USTARA(15,6),R(15,11),IUSEA(11,15,6),
     1 IUSEX(15,6),VDS(15,15,6),VDA(15,15,6),CVDSA(15,6),
     2 USTAR(328,186),VD(15,328,186)
C
C*** Check for beginning and ending of run.
      IF(IFLAG.EQ.0) GOTO 25
      IF(IFLAG.EQ.2) GOTO 30
C
C*** Initialize summation variables.
      DO 20 II=1,15
      DO 20 JJ=1,6
      USTARA(II,JJ)=0.
      CVDSA(II,JJ)=0.
      IUSEX(II,JJ)=0
      DO 15 N=1,11
   15 IUSEA(N,II,JJ)=0.
      DO 20 K=1,15
      VDS(K,II,JJ)=0.
   20 VDA(K,II,JJ)=0.
      IFLAG=0
C
C*** Compute sums.
   25 IF(I.LT.13.OR.I.GT.312) GOTO 100
      IF(J.LT.7) GOTO 100
      II=(I-13)/20+1
      JJ=(J-7)/30+1
      USE=IUSE(LL)
      USE=USE/100.
      IUSEA(LL,II,JJ)=IUSEA(LL,II,JJ)+IUSE(LL)
      USTARA(II,JJ)=USTARA(II,JJ)+
     1USTAR(I,J)*USTAR(I,J)*USE
      CVDSA(II,JJ)=CVDSA(II,JJ)+CVDS*USE
      DO 26 K=1,15
      IF(K.NE.15) VDS(K,II,JJ)=VDS(K,II,JJ)+1./R(K,LL)*USE
   26 VDA(K,II,JJ)=VDA(K,II,JJ)+VD(K,I,J)
      GOTO 100
C
C*** Compute averages.
   30 DO 40 II=1,15
      DO 40 JJ=1,6
      DO 38 LL=1,11
      IUSEA(LL,II,JJ)=IUSEA(LL,II,JJ)/600.
   38 IUSEX(II,JJ)=IUSEX(II,JJ)+IUSEA(LL,II,JJ)
      DO 39 K=1,15
C   Convert surface deposition velocities to resistances.
      IF(VDS(K,II,JJ).NE.0.) VDS(K,II,JJ)=
     1 1./VDS(K,II,JJ)*600.
      IF(VDS(K,II,JJ).EQ.0.)VDS(K,II,JJ)=9999.
   39 VDA(K,II,JJ)=VDA(K,II,JJ)/600.
      IF(CVDSA(II,JJ).NE.0.) VDS(15,II,JJ)=
     1 1./(CVDSA(II,JJ)/600.)
      USTARA(II,JJ)=SQRT(USTARA(II,JJ)/600.)
   40 CONTINUE
C
```

```
C*** OUTPUTS
C*** Write out averaged friction velocities and landuse data.
      WRITE(8,1000)
 1000 FORMAT(' FRICTION VELOCITY (m/s) and LANDUSE COVERAGES
     1 (percent coverage)')
      WRITE(8,1010) ISESN
      WRITE(8,1001)
 1001 FORMAT(' Long.  Lat. USTAR Type: 1   2   3   4   5   6
     1   7   8   9  10  11'/)
      DO 50 II=1,15
      DO 50 JJ=1,6
      IF(IUSEX(II,JJ).LT.90) GOTO 50
      XLONG=52.5+II*5
      XLAT=22.5+JJ*5
      WRITE(8,1002) XLONG,XLAT,USTARA(II,JJ),
     1 (IUSEA(LL,II,JJ),LL=1,11)
   50 CONTINUE
 1002 FORMAT(2F6.1,F6.2,4X,11I4)
      WRITE(8,2000)
 2000 FORMAT('1')
C*** Write out averaged surface res.'s and dep. vel.'s.
C*** First write out values for SO2, SO4, O3, NO3, AND NO.
      WRITE(8,1003)
 1003 FORMAT('    SURFACE RESISTANCES (s/m) and DEPOSITION
     1 VELOCITIES (cm/s)')
      WRITE(8,1010) ISESN
      WRITE(8,1004)
 1004 FORMAT(' Long.  Lat.     SO2          SO4          O3
     1          NO2          NO'/)
      DO 60 II=1,15
      DO 60 JJ=1,6
      IF(IUSEX(II,JJ).LT.90) GOTO 60
      XLONG=52.5+II*5
      XLAT=22.5+JJ*5
      WRITE(8,1005) XLONG,XLAT,VDS(1,II,JJ),VDA(1,II,JJ),
     1 VDS(15,II,JJ),VDA(15,II,JJ),VDS(2,II,JJ),VDA(2,II,JJ),
     2 VDS(3,II,JJ),VDA(3,II,JJ),VDS(4,II,JJ),VDA(4,II,JJ)
   60 CONTINUE
 1005 FORMAT(2F6.1,5(F6.0,F7.3))
      WRITE(8,2000)
C*** Write out values for HNO3, H2O2, ALD, HCHO, and OP.
      WRITE(8,1003)
      WRITE(8,1010) ISESN
      WRITE(8,1006)
 1006 FORMAT(' Long.  Lat.    HNO3         H2O2         ALD
     1         HCHO          OP'/)
      DO 70 II=1,15
      DO 70 JJ=1,6
      IF(IUSEX(II,JJ).LT.90) GOTO 70
      XLONG=52.5+II*5
      XLAT=22.5+JJ*5
      WRITE(8,1005) XLONG,XLAT,VDS(5,II,JJ),VDA(5,II,JJ),
     1 VDS(6,II,JJ),VDA(6,II,JJ),VDS(7,II,JJ),VDA(7,II,JJ),
     2 VDS(8,II,JJ),VDA(8,II,JJ),VDS(9,II,JJ),VDA(9,II,JJ)
   70 CONTINUE
      WRITE(8,2000)
C*** Write out values for PAA, ORA, HN3, PAN, and HNO2.
      WRITE(8,1003)
      WRITE(8,1010) ISESN
      WRITE(8,1007)
 1007 FORMAT(' Long.  Lat.     PAA          ORA          NH3
     1          PAN         HNO2'/)
      DO 80 II=1,15
      DO 80 JJ=1,6
      IF(IUSEX(II,JJ).LT.90) GOTO 80
      XLONG=52.5+II*5
      XLAT=22.5+JJ*5
      WRITE(8,1005) XLONG,XLAT,VDS(10,II,JJ),VDA(10,II,JJ),
     1 VDS(11,II,JJ),VDA(11,II,JJ),VDS(12,II,JJ),VDA(12,II,JJ),
     2 VDS(13,II,JJ),VDA(13,II,JJ),VDS(14,II,JJ),VDA(14,II,JJ)
   80 CONTINUE
      WRITE(8,2000)
 1010 FORMAT('    Seasonal category number',I3/)
      PRINT *,' End of writing out on MODAV.DAT'
  100 CONTINUE
      RETURN
      END
```

```
C    MODINP.DAT
C********** Maximum SO4 dep. velocities (m/s;11F6.3), types 1-11
C       1, Midsummer with lush vegetation
 0.001 0.01  0.01  0.01  0.01  0.01  0.001 0.01  0.01  0.01  0.01
C       2, Autumn with unharvested cropland
 0.001 0.01  0.01  0.001 0.008 0.004 0.001 0.01  0.01  0.01  0.01
C       3, Late autmn after frost, no snow
 0.001 0.01  0.01  0.001 0.008 0.004 0.001 0.01  0.01  0.01  0.01
C       4, Winter with snow on ground
 0.001 0.01  0.01  0.001 0.008 0.004 0.001 0.01  0.01  0.01  0.01
C       5, Transitional spring with partial green coverage
 0.001 0.01  0.01  0.01  0.01  0.01  0.001 0.01  0.01  0.01  0.01
C********** Values of Zo (1.E-4 m; 11I6), landuse types 1-11
C       1, Midsummer with lush vegetation
 10000  2500   500 10000 10000 10000     1    20  1500  1000  1000
C       2, Autumn with unharvested cropland
 10000  1000   500 10000 10000 10000     1    20  1000   800   800
C       3, Late autumn after frost, no snow
 10000    50   500 10000 10000 10000     1    20  1000   200   600
C       4, Winter with snow on ground
 10000    10    10 10000 10000 10000     1    20    10    10   400
C       5, Transitional spring with partial green coverage
 10000   300   200 10000 10000 10000     1    20  1000   300   600
C********** Values of surface resistance components (s/m)
C       1, Midsummer with lush vegetation
ri   9999.   60.  120.   70.  130.  100. 9999. 9999.   80.  100.  150.
rlu  9999. 2000. 2000. 2000. 2000. 2000. 9999. 9999. 2500. 2000. 4000.
rac   100.  200.  100. 2000. 2000. 2000.    0.    0.  300.  150.  200.
rgsS  400.  150.  350.  500.  500.  100.    0. 1000.    0.  220.  400.
rgs0  300.  150.  200.  200.  200.  300. 2000.  400. 1000.  180.  200.
rclS 9999. 2000. 2000. 2000. 2000. 2000. 9999. 9999. 2500. 2000. 4000.
rcl0 9999. 1000. 1000. 1000. 1000. 1000. 9999. 9999. 1000. 1000. 1000.
C       2, Autumn with unharvested cropland
ri   9999. 9999. 9999. 9999.  250.  500. 9999. 9999. 9999. 9999. 9999.
rlu  9999. 9000. 9000. 9000. 4000. 8000. 9999. 9999. 9000. 9000. 9000.
rac   100.  150.  100. 1500. 2000. 1700.    0.    0.  200.  120.  140.
rgsS  400.  200.  350.  500.  500.  100.    0. 1000.    0.  300.  400.
rgs0  300.  150.  200.  200.  200.  300. 2000.  400.  800.  180.  200.
rclS 9999. 9000. 9000. 9000. 2000. 4000. 9999. 9999. 9000. 9000. 9000.
rcl0 9999.  400.  400.  400. 1000.  600. 9999. 9999.  400.  400.  400.
C       3, Late autumn after frost, no snow
ri   9999. 9999. 9999. 9999.  250.  500. 9999. 9999. 9999. 9999. 9999.
rlu  9999. 9999. 9000. 9000. 4000. 8000. 9999. 9999. 9000. 9000. 9000.
rac   100.   10.  100. 1000. 2000. 1500.    0.    0.  100.   50.  120.
rgsS  400.  150.  350.  500.  500.  200.    0. 1000.    0.  200.  400.
rgs0  300.  150.  200.  200.  200.  300. 2000.  400. 1000.  180.  200.
rclS 9999. 9999. 9000. 9000. 3000. 6000. 9999. 9999. 9000. 9000. 9000.
rcl0 9999. 1000.  400.  400. 1000.  600. 9999. 9999.  800.  600.  600.
C       4, Winter with snow on ground
ri   9999. 9999. 9999. 9999.  400.  800. 9999. 9999. 9999. 9999. 9999.
rlu  9999. 9999. 9999. 9999. 6000. 9000. 9999. 9999. 9000. 9000. 9000.
rac   100.   10.   10. 1000. 2000. 1500.    0.    0.   50.   10.   50.
rgsS  100.  100.  100.  100.  100.  100.    0. 1000.  100.  100.   50.
rgs0  600. 3500. 3500. 3500. 3500. 3500. 2000.  400. 3500. 3500. 3500.
rclS 9999. 9999. 9999. 9000.  200.  400. 9999. 9999. 9000. 9999. 9000.
rcl0 9999. 1000. 1000.  400. 1500.  600. 9999. 9999.  800. 1000.  800.
C       5, Transitional spring with partial green coverage
ri   9999.  120.  240.  140.  250.  190. 9999. 9999.  160.  200.  300.
rlu  9999. 4000. 4000. 4000. 2000. 3000. 9999. 9999. 4000. 4000. 8000.
rac   100.   50.   80. 1200. 2000. 1500.    0.    0.  200.   60.  120.
rgsS  500.  150.  350.  500.  500.  200.    0. 1000.    0.  250.  400.
rgs0  300.  150.  200.  200.  200.  300. 2000.  400. 1000.  180.  200.
rclS 9999. 4000. 4000. 4000. 2000. 3000. 9999. 9999. 4000. 4000. 8000.
rcl0 9999. 1000.  500.  500. 1500.  700. 9999. 9999.  600.  800.  800.
```